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Bioenergy production from perennial energy crops: A consequential LCA of 12 bioenergy scenarios including land use changes

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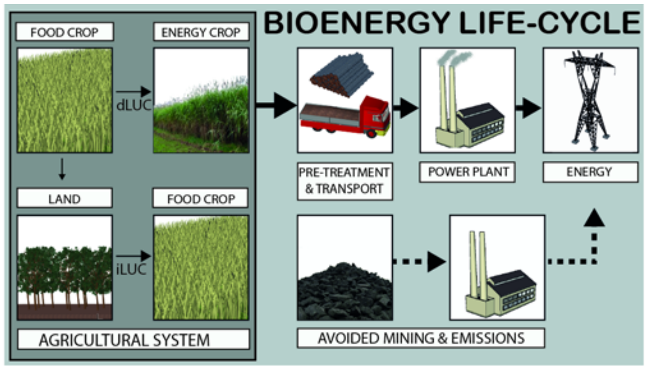

Abstract

In the endeavor of optimizing the sustainability of bioenergy production in Denmark, this consequential life cycle assessment (LCA) evaluated the environmental impacts associated with the production of heat and electricity from one hectare of Danish arable land cultivated with three perennial crops: ryegrass (*Lolium perenne*), willow (*Salix viminalis*) and *Miscanthus giganteus*. For each, four conversion pathways were assessed against a fossil fuel reference: I) anaerobic co-digestion with manure, II) gasification, III) combustion in small-to-medium scale biomass combined heat and power (CHP) plants and IV) co-firing in large scale coal-fired CHP plants. Soil carbon changes, direct and indirect land use changes as well as uncertainty analysis (sensitivity, MonteCarlo) were included in the LCA. Results showed that global warming was the bottleneck impact, where only two scenarios, namely willow and *Miscanthus* co-firing, allowed for an improvement as compared to the reference (-82 and -45 t CO_2-eq. ha^{-1}, respectively). The indirect land use changes impact was quantified as 310 ±170 t CO_2-eq. ha^{-1}, representing a paramount average of 41% of the induced greenhouse gas emissions. The uncertainty analysis confirmed the results robustness and highlighted the indirect land use changes uncertainty as the only uncertainty that can significantly change the outcome of the LCA results.

1. Introduction

The ambition of the energy policy in Denmark is to reach a 100% renewable energy system by 2050 (1). Several studies have been conducted to design and optimize such a system, and these all highlight the indispensability of a biomass potential of around 35%–50% of the overall energy consumption (2-5). There are several reasons explaining why biomass is so attractive for energy systems entirely free of fossil energy (6). Its key advantage, however, lies in the fact that it is storable, entitling it to be used for balancing the fluctuating energy production from intermittent sources like wind and solar power (1, 2, 6, 7).

Though biomass is a renewable energy source, it is not unlimited in supply, and does involve considerable environmental costs. One of the most critical costs of bioenergy relates to its incidence on land use changes (LUC) (8-10), i.e. the conversion of land from one use (e.g. forest, grassland or food/feed crop cultivation) to another use (e.g. energy crop cultivation).

One way to minimize these LUC impacts could be through favouring the cultivation of perennial energy crops (e.g. perennial ryegrass, willow and *Miscanthus*) instead of annual crops (e.g. maize, barley, wheat, sugar beet). In fact, it is acknowledged that perennial energy crops nowadays represent the most efficient and sustainable feedstock available for bioenergy production in temperate regions (11-13). Among others, perennial energy crops generally present a more efficient nutrient use than their annual counterpart, which involves lower requirements for annual inputs of fertilizers, and consequently lower environmental impacts related to fertilization (14). Moreover, in contrast to annual crops whose cultivation tends to accelerate the depletion of soil organic carbon (SOC), perennial energy crops allow for an accumulation of SOC (14). They generally also present higher yields, involve less soil disturbances due to their longer life cycle duration, and have a better incidence on biodiversity (12). For these reasons, this study focuses on bioenergy production from perennial energy crops only.

The goal of this study is to assess the environmental impacts associated with the production of bioenergy (heat and electricity) from 1 hectare (ha) of Danish arable land cultivated with ryegrass, willow and *Miscanthus*, considering four different biomass-to-energy (BtE) conversion pathways: i) anaerobic co-digestion with manure, ii) gasification, iii) combustion in small-to-medium scale biomass combined heat and power (CHP) plants and iv) co-firing in large scale coal-fired CHP plants.

2. Material and methods

2.1 Life cycle assessment model

2.1.1 Scope and functional unit

The environmental assessment presented in this study was performed using consequential life cycle assessment (LCA) (15, 16). The functional unit upon which all input and output flows were expressed was 1 ha of agricultural land used to grow the selected energy crops. The geographical scope considered for the LCA was Denmark, i.e. the data inventory for crops cultivation and BtE plants were specific for Danish conditions. Similarly, the legislative context of Denmark (e.g. fertilization) was considered. The temporal scope considered was 20 years, i.e. all assessed systems were operated for 20y duration.

2.1.2 Impact assessment

The life cycle impact assessment was carried out according to the Danish EDIP 2003 method (17, 18) for the environmental impact categories global warming (aggregated emissions over a 100 years horizon) (GW) and aquatic eutrophication (distinguishing between nitrogen and phosphorus being the limiting nutrient for growth) (EP (N) and EP (P), respectively). To this, an impact category named "Phosphorous as resource" was added, in order to reflect the benefits associated with phosphorous (P) savings, based on the Impact 2002+ method (19). Background LCA data were based on the Ecoinvent v.2.2 database, and the assessment was facilitated by the LCA software SimaPro 7.3.3 (20). Foreground LCA data essentially included Danish-specific data for agricultural and energy conversion processes, and the impacts associated with capital goods (foreground data only) as well as those related to transportation of the residues (i.e. ash and digestate) have been excluded.

2.2 Scenarios modeling and system boundary

The systems assessed considered three perennial crops (ryegrass, willow and *Miscanthus*) and four BtE conversion technologies (anaerobic co-digestion, gasification, combustion in small-to-medium scale biomass CHP plants and co-firing in large scale coal-fired CHP plants). A total of 12 scenarios have therefore been assessed. The system boundary conditions are illustrated in Figure 1, for the case of ryegrass anaerobic co-digestion. The process flow diagrams for the other scenarios are similar, though the pre-treatments and the flows differ, as shown in Table S2 and Figures S1-S11 of the Supporting Information (SI).

For all BtE technologies, the energy produced was considered to be used for CHP production, thereby substituting the production of marginal heat and power. In the present study, the marginal electricity source was assumed to be from coal-fired power plants conformingly with (21, 22), and the marginal heat from natural gas-based domestic boiler, this being the fuel which is most likely to react to a marginal change in the heat demanded/supplied (23) (further detailed in SI).

As illustrated in Figure 1, the digestate produced from anaerobic digestion was used as a fertilizer (for N, P and K), which avoided marginal mineral N, P and K fertilizers to be produced and used, based on the content of N, P and K of the digestate. The marginal N, P and K fertilizers considered were calcium ammonium nitrate, diammonium phosphate and potassium chloride, respectively, conformingly with (14, 24). Further, based on the model from (24), it was considered that the manure portion used for co-digestion would have otherwise been stored and applied on land, without digestion or other treatment.

The three thermal bioenergy scenarios (i.e. gasification, combustion and co-firing) implied negligible residual unconverted carbon that is found in the bottom ashes, fly ashes and eventual waste water. The bottom ashes were assumed to be used for road construction, substituting for natural aggregates, while the fly ashes were assumed to be utilized for backfilling of old salt mines with negligible environmental impacts (25). Treatment of waste water was not included.

All bioenergy scenarios involved the use of Danish agricultural land in order to grow the energy crops. In a country like Denmark, where 68% of the total land is used for cropland and where policies have been adopted in order to double the forested area (nowadays representing ca. 13% of the total land) (26), very limited conversion from forest or alike nature types is occurring. Most likely, the land needed to grow the energy crops will be taken from actual Danish cropland, involving that one crop cultivated today will be displaced. Such a displaced crop is, in consequential LCA, referred to as the marginal crop. In this study, the marginal crop was assumed to be spring barley, based on (22, 27, 28). Based on the consequential LCA logic, as well as on recent studies (9, 29, 30), this resulting drop in supply of Danish spring barley will cause a relative increase in agricultural prices, which then provide incentives to increase the production elsewhere. Such increased crop production may stem from both increased yield and land conversion to cropland, the latter being also referred to as indirect land use change (iLUC) (9, 29, 30). As illustrated in Figure 1, and as in recent iLUC studies (10, 31, 32), this study included the environmental impacts of the latter only.

Figure 1

2.3 Life cycle inventory (LCI)

2.3.1 Crops

The LCI of all crops was based on a recent Danish consequential LCI (14), which comprises all processes involved during the cultivation stage, up to harvest. This included the tillage activities, liming, propagation (seed, rhizome and cutting production), plant protection, fertilization, sowing/planting, harvest and transport from farm to field. A sandy loam soil has been considered for all crops, as well as precipitations of 964 mm y^{-1}. For *Miscanthus* and willow, the C turnover rate in the topsoil was considered to be reduced by 25% in response to the absence of tillage over many years. For all crops, the fertilization operations were performed in conformity with Danish regulations (33, 34), involving an upper limit for the amount of N to be applied on the field, both as mineral fertilizer and animal slurry.

Based on (14), the life cycle considered for perennial ryegrass (short-term ley), willow and *Miscanthus* plantations were respectively 2y, 21y (6 cuts; 3 years harvest cycle, but first harvest after 4 years; 1 year establishment; 1 year preparation before planting) and 20y (18 cuts; 1 year establishment: 1 year preparation before planting). Given the 20y temporal scope of the LCA, this means that the life cycle of ryegrass, willow and *Miscanthus* is respectively occurring 10, 0.95 and 1 time. Further, it was considered that ryegrass was harvested in summer, willow in the vegetative rest period (in the period around November to February) and *Miscanthus* during the spring season.

2.3.2 BtE conversion technologies and pre-treatments

Anaerobic digestion was modelled as mesophilic co-digestion of the respective energy crops with raw pig manure. Manure represents one of the most abundant domestically available biomass resources in Denmark (ca. 23-34 PJ), which is nowadays significantly underexploited for energy production (5). The current management of raw manure consists to store it in an outdoor structure until it can be used as an organic fertilizer on agricultural land, which leads to large impacts on most environmental compartments, mainly global warming and eutrophication (24). Hence, co-digestion of manure with carbon-rich biomass may represent a viable alternative to produce bioenergy and improve manure management. The modelled methane yields for ryegrass, willow, *Miscanthus* and raw pig manure were, respectively, 290, 240, 250 and 320 $Nm^3\ t^{-1}VS$ (see SI). Based on (24), the mixture of crop and raw pig manure was calculated in order to ensure a biomass mixture input having a dry matter (DM) content of 10% after the first digestion step. The resulting ratio manure:crop (fresh weight basis) for co-digestion of ryegrass, willow and *Miscanthus* equaled 5.7, 6.4 and 6.7, yielding respectively 140, 160 and 130 MJ $CH_4\ ha^{-1}$ (Table S9). Consumption of electricity (2% of the energy in the biogas) and heat (to heat up the substrates from 8 to 37 ºC) was modelled according to (24). Fugitive CH_4 emissions were taken as 1% of the produced CH_4, based on recent studies (24, 35, 36). More details on the modelling of anaerobic digestion can be found in the SI.

Gasification was modelled as fluidized bed gasification based on a number of reviewed studies (Table S5). The resulting cold gas and carbon conversion efficiency (CGE and CCE) was 70% (±15%) and 95% (±4%), respectively. Consumption of electricity (26 kWh $Mg^{-1}DM$) was based on (36).

Combustion was modelled as direct biomass combustion in small-to-medium scale biomass CHP plants, based on a thorough review of (mainly Danish) biomass CHP plants (Table S6). Average net electricity and heat efficiencies inventoried from this review were 27% (±2%) and 63% (±7%), respectively. Co-firing in large scale coal-fired CHP plants was likewise modelled, resulting to net electricity and heat efficiencies of 38% (±3%) and 52% (±8%), respectively (SI).

The air emissions from biogas and syngas combustion in gas engines as well as from biomass combustion in CHP plants were based on (37) (Table S7). Both biogas and syngas were assumed utilized in a gas engine with an average gross electricity and heat efficiency of 38% (±4%) and 52% (±8%) (of the LHV of the input-gas).

Pre-treatments included on field drying (ryegrass, for all BtE conversion technologies) and natural drying (willow, for gasification and co-firing), size comminution (all crops, for all BtE conversion technologies except direct combustion) as well as steam pre-treatment for breaking the lignocellulosic structures of *Miscanthus* and willow undergoing anaerobic digestion. All these pre-treatments are further detailed in the SI.

2.3.3 Other processes

Additional processes modelled in the LCA were: crops and digestate storage, use on land (UOL) of the digestate, treatment of residues from thermal BtE technologies and transportation. A detailed description of these processes can be found in the SI.

2.4 Carbon and nitrogen flow analysis

Carbon and nitrogen flows are two of the most important flows responsible for the environmental impacts involved in bioenergy systems. Therefore, the C and N flows of all the scenarios assessed in this study have been disaggregated and calculated for all the major processes involved. This included the soil C changes resulting from the cultivation stage, which were calculated with the dynamic soil C model C-TOOL (38, 39), as detailed in (14) for all crop systems. The modeling of the other C and N flows was based on the equations listed in the SI. The carbon and nitrogen flow analysis was facilitated by the software STAN (40) allowing a quantification of the uncertainties for the most sensitive parameters (Table S17) and to reconcile the data when necessary.

2.5 Direct and Indirect land use changes impacts

As earlier explained, the LCA system established in this study considers that the land used for cultivating the energy crops would have otherwise been used for cultivating spring barley (with straw incorporation) for the food/feed market (Figure 1). The direct land use change (dLUC) consequence of this translates into the environmental impacts of cultivating the selected energy crops instead of spring barley (Figure 1). The environmental impacts from spring barley cultivation have been included on the basis of the data from (14).

The iLUC consequence corresponds to the environmental impact of converting land nowadays not used for crop cultivation to cropland, as a result of the induced demand for the displaced spring barley. To quantify this impact, it is necessary to identify i) how much land is converted and where; and ii) which types of land are converted (biome types). So far, most studies attempting to quantify the magnitude of iLUC used econometric models to this end, e.g. (9, 10, 29, 31, 32), where the economic and biophysical/agricultural systems are combined into one single modeling framework. A comprehensive overview of partial and general equilibrium models that can be used to model iLUC is given in (41).

Most of available iLUC studies to date focused on biofuel mandates for a variety of shock sizes, and as such are difficult to be used directly for other applications. In (29), however, the iLUC consequences in terms of points i) and ii) above are identified, for a marginal increase in wheat consumption in 4 different countries, including Denmark. This was done using a modified version of the general equilibrium GTAP model (42). In the present study, the results of (29) for Denmark have been used as a proxy to estimate how much land is converted (due to the increased spring barley demand) and where. However, the CO_2 impact of land conversion is not estimated in (29). In order to do so, the soil and vegetation C data from the Woods Hole Research Centre, as published in (9), have been used, and the CO_2 emitted due to land conversion was calculated based on the methodology published in (43). Based on this methodology, it was considered that 25% of the C in the soil was converted to CO_2 for all types of land use conversion, except when forests were converted to grassland, where 0% was converted. Further, it was considered that 100% of the C in vegetation was converted to CO_2 for all forest types as well as for tropical grassland conversions, while 0% was converted for the remaining biome types (e.g. shrub land, non-tropical grassland, chaparral).

2.6 Sensitivity and uncertainty analysis

Two types of uncertainties were addressed in this study (for the GW impact only), namely scenario and parameter uncertainties. While the former deals with the uncertainty due to the intrinsic modeling choices (in terms of system boundary and marginal technologies/products), the latter covers the uncertainty related to the quantification of the values used in the LCA model.

Parameter uncertainties were addressed through a MonteCarlo analysis (number of simulations: 1000), whereas scenario uncertainties were addressed through sensitivity analyses. These included: a) variation (min-max) of the iLUC impacts with respect to CO_2 emissions (vs. mean value assumed as baseline); b) winter wheat as the marginal crop for Denmark (vs. spring barley as baseline); c) coal-based heat production as the marginal technology for heat generation (vs. natural gas-based as baseline); d) natural gas power plant as the marginal technology for electricity generation (vs. condensing coal power plant as baseline); e) mono-digestion of the crops (vs. co-digestion with manure as baseline); f) pre-treatment of pelletization before co-firing (vs. 'no pelletization' as baseline). Each of these changes was tested individually to assess the influence of the individual change on the overall LCA results.

A thorough description of the methodology used for sensitivity and uncertainty analysis can be found in the SI.

3. Results and Discussion

3.1 Carbon and nitrogen flows

The induced C and N flows for ryegrass, willow and *Miscanthus* are presented in Figures S13-S18 (SI).

As illustrated in Figures S13-S15, more than 85% of the C input to the energy crop system (the most notable being the uptake from the atmosphere) ends up emitted as CO_2, whether as a result of the cultivation stage or as a result of the final energy use. As indicated in (8, 44), many bioenergy studies report rather different results, as the biogenic CO_2 emissions from the cultivation stage (releases from manure and residues not entering the soil C pool), which here represents 39%-45% (Table S8) of the C input fate, are not accounted for. This highlights the importance of the error made if a complete system-based mass balance, such as the one performed in this study, is not considered.

The C from atmospheric uptake was similar for all the three crops (about 11-12 t C $ha^{-1}y^{-1}$): for all crops, only about half of this C ended up in the harvested biomass, the other half ending up in the non-harvested above- and below-ground residues (Figures S13-S15). The biogenic CO_2 emission related to crop cultivation (6.1 to 6.9 t CO_2-C $ha^{-1}y^{-1}$) was also in the same order of magnitude for all crops (Figure S13-S15; Table S8). The biogenic carbon emission from the final energy use, however, varied significantly more (2.9 to 6.0 t CO_2-C $ha^{-1}y^{-1}$), as detailed in Table S8. This reflects the importance of two main parameters: the crop yield and the BtE technology. In fact, the biogenic CO_2 emission from the final energy use was the greatest for thermal treatments (combustion and gasification), where 95%-100% of the carbon was emitted as CO_2, whereas it was significantly lower for biological treatment (anaerobic co-digestion), where only ca. 41%-56% of the crop (and raw manure) carbon was gasified (Table S8). This unconverted C during anaerobic co-digestion is ultimately applied on land, through the digestate. However, this did not represent a significant carbon sink, as more than two thirds of this C was released as CO_2, rather than sequestrated in the soil (Figure S13-S15). This is in accordance with previous findings (e.g. (24)).

The variation in SOC due to dLUC was positive (i.e. the SOC content was increased) for all crop systems. This was expected, since spring barley, an annual crop with a much lower yield than any of the perennial energy crops considered here, involves losses instead of gains in soil C, as illustrated in (14). The modeled ΔSOC was very similar for the three crops (about 0.7 t C $ha^{-1}y^{-1}$). The avoided CO_2 emissions resulting from the substitution of fossil carbon were proportional to the amount of bioenergy produced; this ranged from 3.9 (anaerobic digestion of *Miscanthus*) to 8.3 (co-firing of willow) t C $ha^{-1}y^{-1}$ (Table S8).

As opposed to C, the outputs of N flows were more diversified among the individual flows. The most significant N flows occurred during the UOL of the digestate for the anaerobic co-digestion scenarios, and during the cultivation stage for the other scenarios (Figures S16-S18; Table S8). Ryegrass showed the highest emissions of N during the cultivation phase; these occurred as a consequence of the higher nitrogen fertilizer requirements of ryegrass (450 kg N $ha^{-1}y^{-1}$) compared to willow (170 kg N $ha^{-1}y^{-1}$) and *Miscanthus* (70 kg N $ha^{-1}y^{-1}$). These fertilization rates (and the related N-based emissions) are based on today's practices, but should be seen as reflecting the highest end of the interval. In fact, *Miscanthus* and willow are relatively new crops, and it can be expected that lower application rates will be required as insight is gained on the optimal management of these crops (45, 46). Similarly, lower N application could be considered for ryegrass dedicated to bioenergy, where protein production is not the focus (as in the case of forage ryegrass). The N-related emissions at the UOL stage (anaerobic co-digestion scenarios) were similar for all the three crops, as a consequence of the Danish legislation for fertilization fixing the maximal amount of N to be applied in agricultural fields (33, 34). Overall, NO_3^- and NH_3 emissions were the most significant N-emissions.

3.2 Indirect land use changes

The iLUC impacts of the studied bioenergy systems were the same for all scenarios (Figure 2a), as they all had the same "point of origin": the conversion of 1 ha of Danish land (cultivated with spring barley) to energy crops. As shown in Table 1 (and further detailed in the SI), these iLUC impacts were estimated to 310 t CO_2-eq. ha^{-1} (± 170 t CO_2-eq. ha^{-1}). The impacts were annualized over a period of 20 years in accordance with IPCC (47) and with prominent European legislation (48), corresponding to about 16 t CO_2-eq. $ha^{-1}y^{-1}$ (or 70-130 g CO_2-eq. $MJ^{-1}y^{-1}$).

Table 1

Although currently debated and relatively uncertain (49), the iLUC impact quantified here can contribute with important learnings: i) it is not zero; and ii) it may cover a significant proportion of the overall global warming impact (Figure 2a) (between one third to half of the positive contributions, depending on the scenario), and cancels out the otherwise avoided GHG emissions in the scenarios. Moreover, it should be highlighted that the 310 t CO_2-eq. ha^{-1} obtained here only covers the GHG related to the net expansion resulting from the modeling of (29) and does not include the GHG related to the intensification of crop production (which accounts, based on the results of (29), to about 30% of the displacement response). This suggests that the "real" impact may actually be higher. The only other LCA study (50) the authors were aware of attempting to quantify iLUC on the basis of an hectare of land displaced (and not a biofuel mandate shock) led to a considerably higher value, i.e. 440-560 t CO_2-eq. ha^{-1} (considering a 20 years period and only conversion of forest). Although it cannot be directly compared, our annualized iLUC value (70-130 g CO_2-eq. $MJ^{-1}y^{-1}$, calculated dividing the annualized iLUC impact by the energy yielded by 1 hectare cultivated with the crops, dry basis) lies within the range of values found in (10) for marginal increases in the demand for biofuels.

In this study, the assessment of global warming was based on the IPCC AR4 methodology (51), where GHG are summed up over a defined time horizon, which in LCA is commonly taken as 100y (as in this study). The use of this approach may however be seen as a limitation when emission releases occurring at different times (e.g. year 0 and year 13) are involved, as these releases are then summed together despite that their end points of analysis are different (e.g. year 100 and year 113). In recent years, a number of studies have proposed methodologies to address this flaw, where many emphasized the particular case of iLUC (e.g. (43, 52, 53)). As these methodologies are still in their early development stage, the global warming results presented in this study are based on the IPCC methodology. However, the importance of time-dependency was assessed for the cultivation of *Miscanthus* (including iLUC), based on the methodology described in (53) (SI). This specific simulation indicated that accounting for time-dependency would increase our GWP by ca. 40%. Such increase was also suggested by the results of (52), for a different bioenergy case.

3.3 LCA results

The environmental impacts related to the 12 bioenergy scenarios assessed are shown in Figure 2 for the selected impact categories. Impacts/savings for the individual bioenergy scenarios were obtained by subtracting the avoided impacts (negative values in the figures) from the induced impacts (positive values). The zero axis represents the reference: any net value below the zero axis thus indicates an environmental improvement as compared with the fossil fuel reference (in which: electricity and heat are provided by coal and natural gas, the hectare of land is used for spring barley cultivation, and manure is not digested).

On the selected impact categories, global warming appears critical as only two scenarios indicate overall savings for this category as compared with the fossil fuel reference. Only co-firing of willow and *Miscanthus* indicated net overall savings, i.e. these were the only two scenarios for which an environmental benefit, GHG-wise, was identified in relation to using 1 ha of land for bioenergy. However, the magnitude of the global warming impacts found in this study (between -82 and 270 t CO_2-eq. ha^{-1} over 20 years) was much higher than previous results from literature. For instance, (54) calculated a saving between -18 and -35 t CO_2-eq. $ha^{-1}y^{-1}$ (-360 to -700 t CO_2-eq. ha^{-1} over 20 years) for bioenergy systems based on willow and *Miscanthus* plantations in Ireland; (55) quantified savings of -25 t CO_2-eq. $ha^{-1}y^{-1}$ (about -500 t CO_2-eq. ha^{-1} in 20 years) for bioenergy systems based on *Miscanthus* plantations in Italy; (56) estimated a saving between -10.4 and -11.1 t CO_2-eq. $ha^{-1}y^{-1}$ (-210 to -220 t CO_2-eq. ha^{-1} in 20 years) for *Miscanthus* and willow plantations in the UK. The reason for these differences is that this study, as opposed to the previous, considered iLUC, which has tremendous significance on the overall GHG balance as earlier discussed.

As illustrated in Figure 2a, the 35% GHG emission saving required in the EU Renewable Energy Directive (48) for biofuels and bioliquids (as compared with the same energy provided from fossil fuels) has been used as a comparative measure of the GHG reductions achieved in the individual scenarios (although the directive does not apply to these scenarios), see calculation details in the SI. As shown in Figure 2a, none of the assessed bioenergy scenarios would comply with a 35% GHG reduction target. This highlights the difficulties for bioenergy to compete with fossil fuels for producing heat and power. Though other renewable energy sources (e.g. wind, solar, hydro) should be prioritized, biomass (residual and energy crops) remains needed in a renewable energy system for its intrinsic versatility (2-5). In this perspective and in the light of Figure 2a, co-firing or efficient combustion of willow and *Miscanthus* can be highlighted as preferable options for producing bioenergy from perennial crops, both in relation to global warming but also to the other impact categories assessed (aquatic P and N eutrophication, P savings).

Co-firing and combustion provided the smallest global warming impacts for all crops. The environmental performance of co-firing was directly related to the higher electricity efficiency of these plants (about 38% of the LHV of the fuel, wet basis), and consequently to the larger amount of marginal coal electricity substituted. Co-firing of willow provided the largest savings, mostly because of the beneficial dLUC, higher yield and minimal pre-treatment required. Similarly, the environmental performance of combustion was due to the high overall energy recovery as heat and electricity (about 90% of the LHV of the fuel, wet basis). As opposed to combustion and co-firing, anaerobic digestion and gasification involved a conversion to gas before energy generation, thereby inducing additional losses (Table S9). Therefore, less electricity and heat were produced and substituted, resulting in larger net GW impacts from these technologies. Further, UOL of the digestate contributed with a GW impact comparable to the one of iLUC, i.e. ranging between 280 (*Miscanthus*) and 370 (willow) t CO_2-eq. ha^{-1}, primarily connected to the release of biogenic carbon not entering the soil C pool (quantified in Figure S13-S15 of the SI). This cannot be directly visualized on Figure 2a, which presents the net impact of UOL (digestate minus raw manure). Co-digestion also resulted in GHG savings associated with avoiding raw manure management, which would otherwise be stored and applied on land without digestion (24). These savings depended on the amount of manure co-digested (per hectare), i.e. the more manure co-digested (to meet the 10% DM in the input-mixture), the larger the savings were. This also applied for aquatic N-eutrophication, where the impacts were much higher for ryegrass because of the higher N content of the crop.

Figure 2 highlights the significance of dLUC for all scenarios and impact categories, where changing from spring barley to perennials generally resulted in environmental benefits. For global warming, this reflects two main points. First, that the perennial crops considered in this study have a much greater C uptake than spring barley. Second, that they are also more efficient systems for converting the C uptake to useful C (i.e. more C in the harvested biomass, less C in the residues, therefore less C lost as CO_2 emissions during the cultivation stage). For the other impact categories, the dLUC results for ryegrass differed from those of *Miscanthus* and willow. Figure 2b for example reflects the high load of N fertilizers applied in the ryegrass system, which resulted in much higher N leaching than in the reference (barley cultivation), while willow and *Miscanthus* systems resulted in a dLUC improvement. On the other hand, as half of the N fertilizers used during cultivation came from animal slurries (14) (which also contain P), no mineral P fertilizers needed to be applied for ryegrass, as opposed to all other crop systems, which explains the greater P savings for this crop in connection with dLUC (Figure 2d). It should however be kept in mind that the high N-leaching results for ryegrass should be seen as a maximum, as ryegrass-for-bioenergy likely requires less N than ryegrass-for-fodder in order to reach the same yields as considered in this study.

In Figure 2d, the category "others" reflects the net induced P fertilizers: since fertilization is by law based on crops N balance (33, 34), even though anaerobic co-digestion allows for nutrients recycling, the higher nutrients content of the produced digestate involves that relatively more P was applied in excess in the co-digestion scenarios compared to the reference (use on land of raw pig manure), thus decreasing the overall P-saving potential and increasing leaching (Figure 2c). P-leaching was less for willow as a consequence of the lower P content of the crop.

Figure 2

The results of the sensitivity analyses highlighted that the variation of the iLUC impacts played the most important role for GW; with minimum iLUC impacts (Table 1) all bioenergy scenarios for willow and *Miscanthus* as well as co-firing of ryegrass achieved environmental savings on GW (Figure S19). Co-firing and combustion of willow and *Miscanthus* even reached the 35% GHG reduction target. In all other analyses, the individual changes in assumptions did not alter the conclusions relative to the baseline. However, the different assumptions made regarding marginal energy and crop decreased or increased the magnitude of the impacts or savings in all scenarios (Figure S19). In the case of mono-digestion, GW impacts were significantly increased as compared to their levels in the co-digestion scenarios (increase between 110 and 160 t CO_2-eq. ha^{-1}), reflecting the tremendous benefits obtained when avoiding conventional manure management. Co-digestion with manure shall therefore be favored in order to optimize the GW savings associated with anaerobic digestion. The sensitivity analysis also demonstrated that additional pelletization and milling of the biomass in the co-firing scenarios would decrease the GW performance of these scenarios to a level very close to direct biomass combustion. The results of the MonteCarlo simulation for GW (Table S18) supported the ranking of the bioenergy scenarios found with the baseline scenarios, demonstrating that despite the significant uncertainties, the results obtained were robust. For gasification, combustion and co-firing, it also highlighted that it was not clear whether the willow scenarios really yielded greater savings than the *Miscanthus* scenarios.

Overall, co-firing of *Miscanthus* and willow appeared to be the options with the best environmental performance. It should however be realized that a main driver for future utilization of biomass may be to balance electricity generation from fluctuating energy sources, such as wind and solar power. Not all biomass combustion technologies may be suited for this, especially when co-generation of heat is important as such plants can have a fixed production ratio between electricity and heat. Anaerobic co-digestion as well as gasification of biomass, on the other hand, may be operated more flexible without similar constraints. Additionally, syngas or biogas offers the flexibility of storage. On this basis, improving the environmental performance of these BtE conversion technologies would be desirable. For anaerobic digestion, a solution may be to favor manure-based biogas together with co-substrates not involving iLUC (e.g. straw, organic municipal household waste, garden waste) as well as in boosting the digestion process by other means (e.g. digestion in series, addition of hydrogen, etc.).

Supporting Information (SI)

Additional information on: marginal energy technologies and fertilizers, LCA process flow diagrams, LCI of crops and BtE conversion technologies, carbon and nitrogen flow charts, energy balance, GWP time-dependency, iLUC and modelling equations as well as sensitivity and uncertainty analyses is available free of charge via the Internet at http://pubs.acs.org.

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Tables

Table 1 Estimation of the iLUC CO_2 impact[*].

Biomes converted[**]	Type of conversion[†]	Region[†‡]	$m^2\ t^{-1}$ wheat[†φ]	C in vegetation ($t\ ha^{-1}$)[β]	C in soil ($t\ ha^{-1}$) [β]	CO_2-C lost ($t\ C\ t^{-1}$ wheat)[∂]	CO_2 lost ($t\ CO_2\ t^{-1}$ wheat)	CO_2 lost ($t\ CO_2\ ha^{-1}$)[λ]
Savanna (taken as shrub land)	100% cropland	xss	140 ± 86	4,6	30	0.11 ± 0.06	0.39 ± 0.24	2.2 ± 1.3
African tropical evergreen forest (taken as tropical rain forest)	100% cropland	xss	140 ± 86	130	190	2.5 ± 1.5	9.1 ± 5.5	52 ± 31
Open shrubland (taken as shrub land)	100% grassland	xss	81 ± 49	4,6	30	0.06 ± 0.04	0.22 ± 0.13	1.3 ± 0.8
Temperate evergreen forest	100% cropland	xeu15	57 ± 34	160	130	1.1 ± 0.7	4.0 ± 2.4	23 ± 14
Temperate deciduous forest	100% cropland	xeu15	57 ± 34	120	130	0.87 ± 0.52	3.2 ± 1.9	18± 11
Dense shrub land (taken as temperate grassland)	46% cropland; 54% grassland	xeu15	250 ± 148	7,0	190	1.2 ± 0.7	4.3 ± 2.6	24 ± 15
Tropical evergreen forest	100% cropland	bra	180 ± 70	200	98	4.0 ± 1.6	15 ± 6	83 ± 33
Savanna (taken as grassland)	100% grassland	bra	41 ± 16	10	42	0.04 ± 0.02	0.16 ± 0.06	0.91 ± 0.36
Grassland/steppe (taken as temperate grassland)	100% cropland	xsu	91 ± 55	10	190	0.43 ± 0.26	1.6 ± 0.9	9.0 ± 5.4
Temperate evergreen forest	100% grassland	xsu	45 ± 27	160	130	0.88 ± 0.43	3.2 ± 1.6	18.3 ± 9.1
Temperate deciduous forest	100% grassland	xsu	45 ± 27	140	130	0.76 ± 0.37	2.8 ± 1.3	16 ± 8
Savanna (taken as tropical grassland)	100% cropland	aus	110 ± 64	18	42	0.31 ± 0.18	1.1 ± 0.7	6.4 ± 3.8
Open shrubland + grassland/steppe (taken as tropical grassland)	100% grassland	aus	37 ± 22	18	42	0.11 ± 0.06	0.39 ± 0.23	2.2 ± 1.3
Boreal deciduous forest (taken as temperate deciduous forest)	100% cropland	can	97 ± 58	140	130	1.6 ± 1.0	6.0 ± 3.6	34 ± 20
Boreal evergreen forest (taken as temperate evergreen forest)	100% grassland	can	10 ± 6	160	130	0.16 ± 0.10	0.59 ± 0.35	3.3 ± 2.0
Grassland/steppe (taken as grassland)	100% cropland	xla	35 ± 21	10	42	0.04 ± 0.02	0.14 ± 0.08	0.77 ± 0.46
Tropical evergreen forest	100% cropland	xla	35 ± 21	200	98	0.79 ± 0.48	2.9 ± 1.7	17 ± 10
Savanna + dense shrub land (taken as grassland)	100% grassland	xla	16 ± 10	10	42	0.02 ± 0.01	0.063 ± 0.038	0.36 ± 0.22
Open shrub land (taken as chaparral)	100% grassland	usa	68 ± 41	40	80	0.14 ± 0.08	0.50 ± 0.30	2.8 ± 1.7
TOTAL	-	-	1500 ± 880	-	-	15 ± 8	54 ± 30	310 ± 170

* Eventual inconsistencies due to rounding (numbers are reported with 2 significant digits).
** Indicated biomes are as in (29). When the biomes mentioned in (29) did not figure in the biomes from the Woods Hole Research Centre data (9), an equivalent was considered, which is indicated between parentheses, when it applies.
† Based on the results from (29).

‡ With xss: Sub-Saharan Africa, excluding Botswana, Lesotho, Namibia, South Africa and Swaziland; xeu15: EU-15, excluding Denmark; bra: Brazil; xsu: Former Soviet Union, excluding the Baltic States; aus: Australia; can: Canada; xla: South America, excluding Brazil and Peru; usa: United States. As indicated in (29), this aggregation covers 92% of the total net expansion.

β From the Woods Hole Research Centre, as published in (9).

∂ Considering that 25% of the C in soil is converted, for all biomes, except when forest is converted to grassland, where 0% of soil C is converted; 100% of the C in vegetation is converted for all forest biomes; 100% of the C in vegetation is converted for tropical grasslands; 0% of the C in vegetation is converted for all other biomes.

λ The conversion per ha is made considering that it is 1 ha of spring barley that is initially displaced, with a yield of 4.9 t DM ha^{-1} and a DM content of 85% of the crop fresh matter, based on (14).

φ The maximal and minimal range are based on the qualitative description of the uncertainty related to the biomes conversion results made by (29). The levels identified as "very good", "good" and "moderate" were considered as an uncertainty of ±20%, 40% and 60%, respectively.

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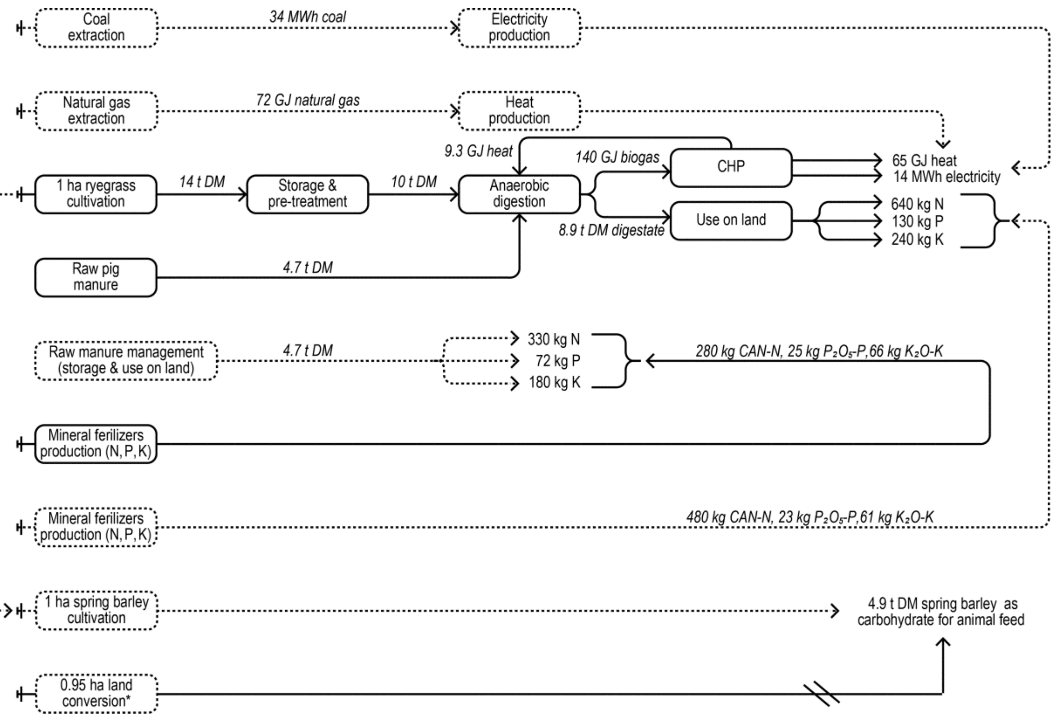
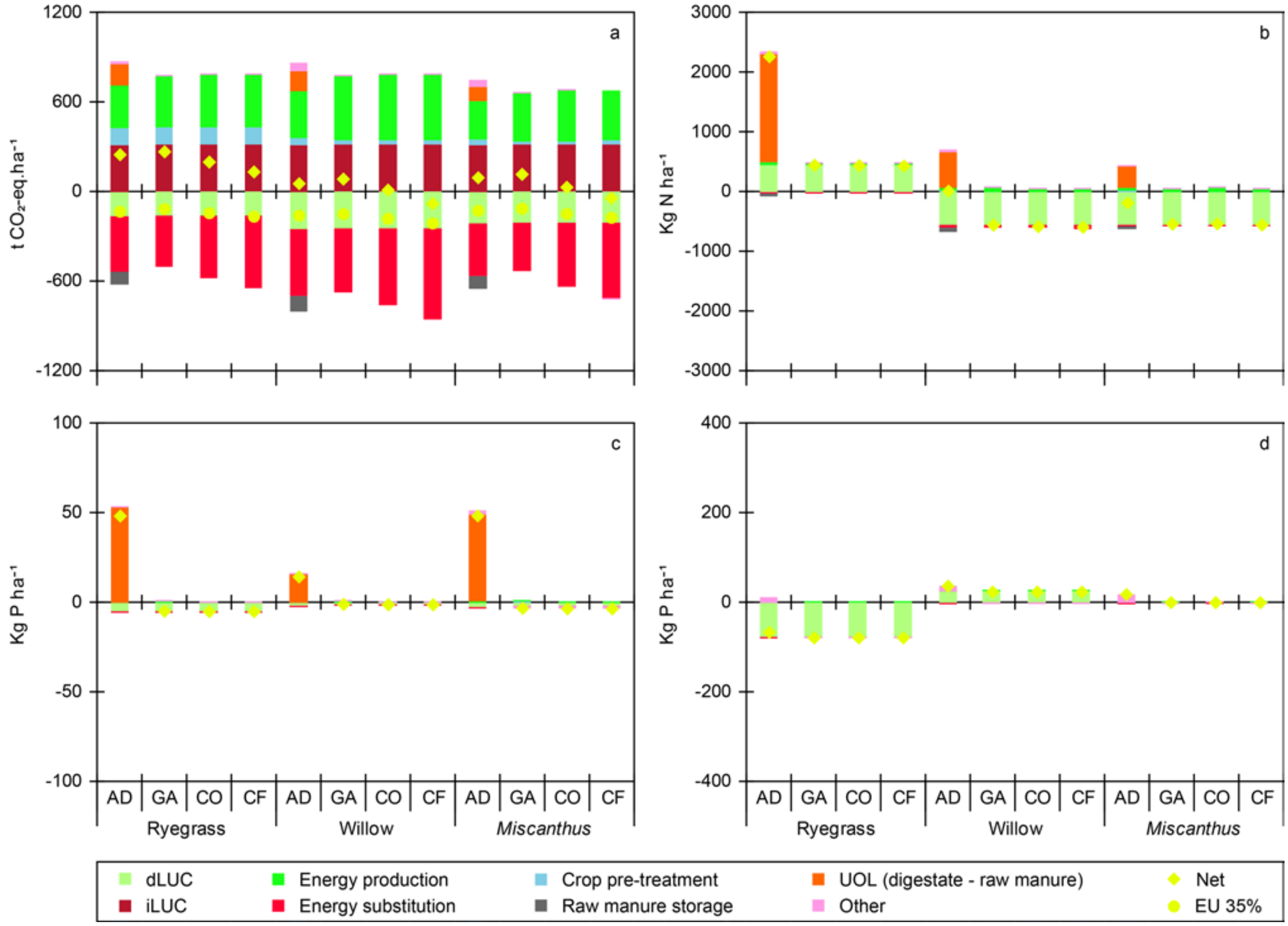


Coal extraction
34 MWh coal
Electricity production
Natural gas extraction
72 GJ natural gas
Heat production
9.3 GJ heat
140 GJ biogas
CHP
65 GJ heat
14 MWh electricity
1 ha ryegrass cultivation
14 t DM
Storage & pre-treatment
10 t DM
Anaerobic digestion
Use on land
8.9 t DM digestate
640 kg N
130 kg P
240 kg K
Raw pig manure
4.7 t DM
Raw manure management (storage & use on land)
4.7 t DM
330 kg N
72 kg P
180 kg K
280 kg CAN-N, 25 kg P_2O_5-P,66 kg K_2O-K
Mineral ferilizers production (N, P, K)
Mineral ferilizers production (N, P, K)
480 kg CAN-N, 23 kg P_2O_5-P,61 kg K_2O-K
1 ha spring barley cultivation
4.9 t DM spring barley as carbohydrate for animal feed
0.95 ha land conversion*



a
b
c
d
t CO2-eq.ha-1
Kg N ha-1
Kg P ha-1
Kg P ha-1
1200
600
0
-600
-1200
3000
2000
1000
0
-1000
-2000
-3000
100
50
0
-50
-100
400
200
0
-200
-400
AD GA CO CF AD GA CO CF AD GA CO CF
Ryegrass
Willow
Miscanthus
dLUC
iLUC
Energy production
Energy substitution
Crop pre-treatment
Raw manure storage
UOL (digestate - raw manure)
Other
Net
EU 35%

Supporting Information (SI) for:

Bioenergy production from perennial energy crops: a consequential LCA of 12 bioenergy scenarios including land use changes

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This SI document includes text, tables and figures with details on the process data for the inventory analysis of the LCA.

Contents

1. Scenarios modeling and system boundary

As thoroughly described in the main manuscript, the systems assessed considered three perennial crops: ryegrass (*Lolium perenne*), willow (*Salix viminalis*) and *Miscanthus* (*giganteus*) and four energy conversion technologies (anaerobic digestion, gasification, combustion in small-to-medium scale biomass CHP plants and co-firing in large scale coal-fired CHP plants). A total of 12 scenarios have therefore been assessed. For the case of anaerobic co-digestion of ryegrass with raw pig manure, the system modeled as well as the boundary conditions considered are illustrated in Figure 1 of the main manuscript. For the remaining bioenergy scenarios the boundary conditions considered are illustrated in Figure S1-S11 (functional unit: 1 hectare of Danish arable land). Notice that electricity and heat produced are net values (i.e., plants own consumptions have been subtracted).

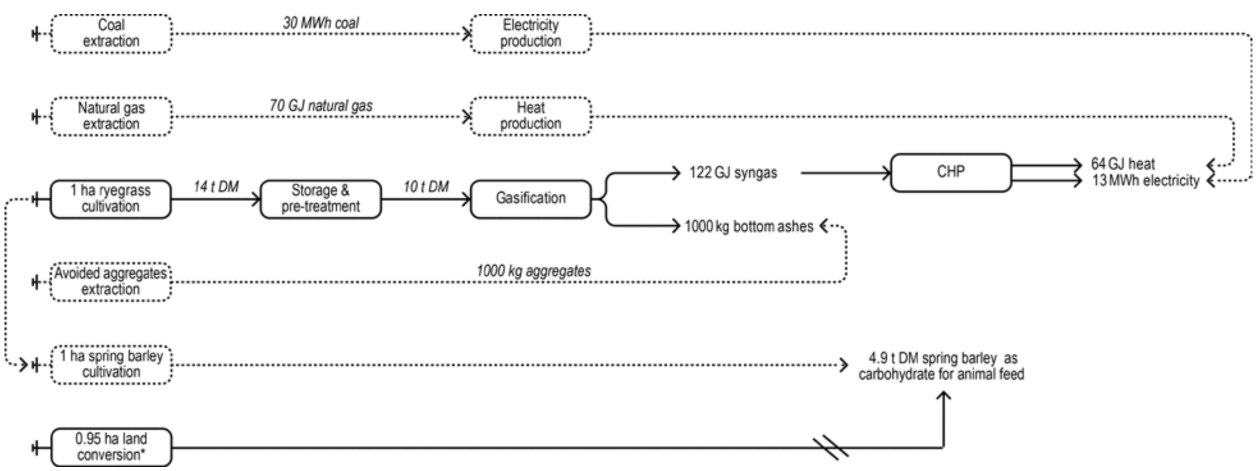

Figure S1. Process flow diagram for gasification of ryegrass. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended). Values are rounded to 2 significant digits.

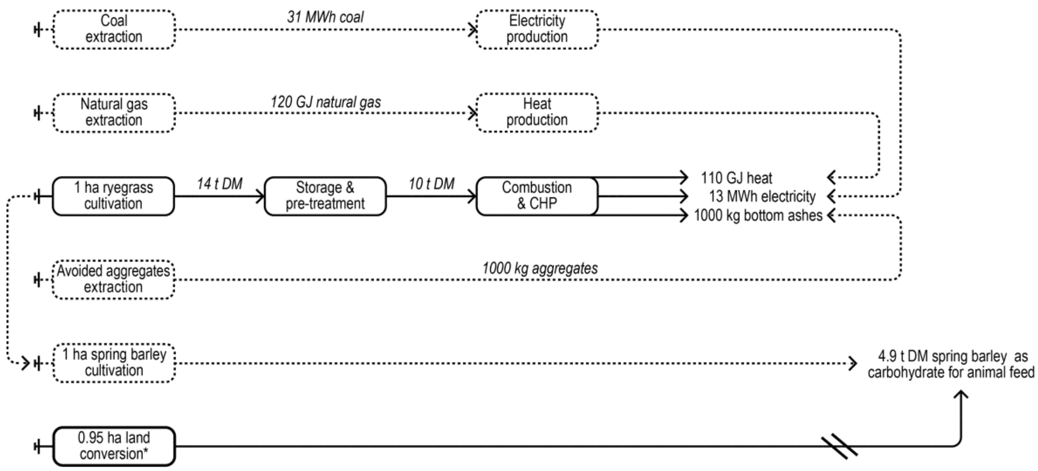

Figure S2. Process flow diagram for combustion of ryegrass. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended). Values are rounded to 2 significant digits.

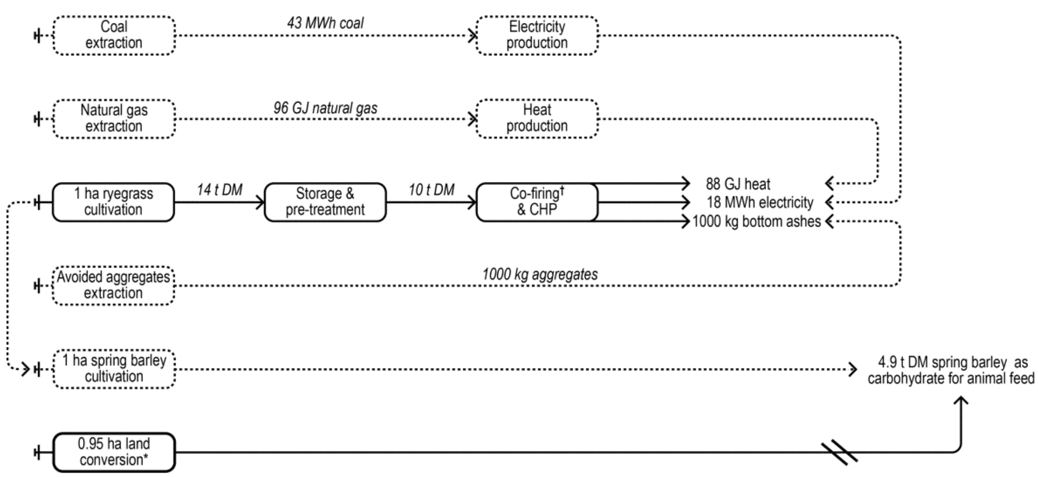

Figure S3. Process flow diagram for co-firing of ryegrass. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Based on the data from co-firing Danish plants, the coal that is used here would have otherwise been used for CHP production, at similar conversion efficiency. Further, fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended). Values are rounded to 2 significant digits.

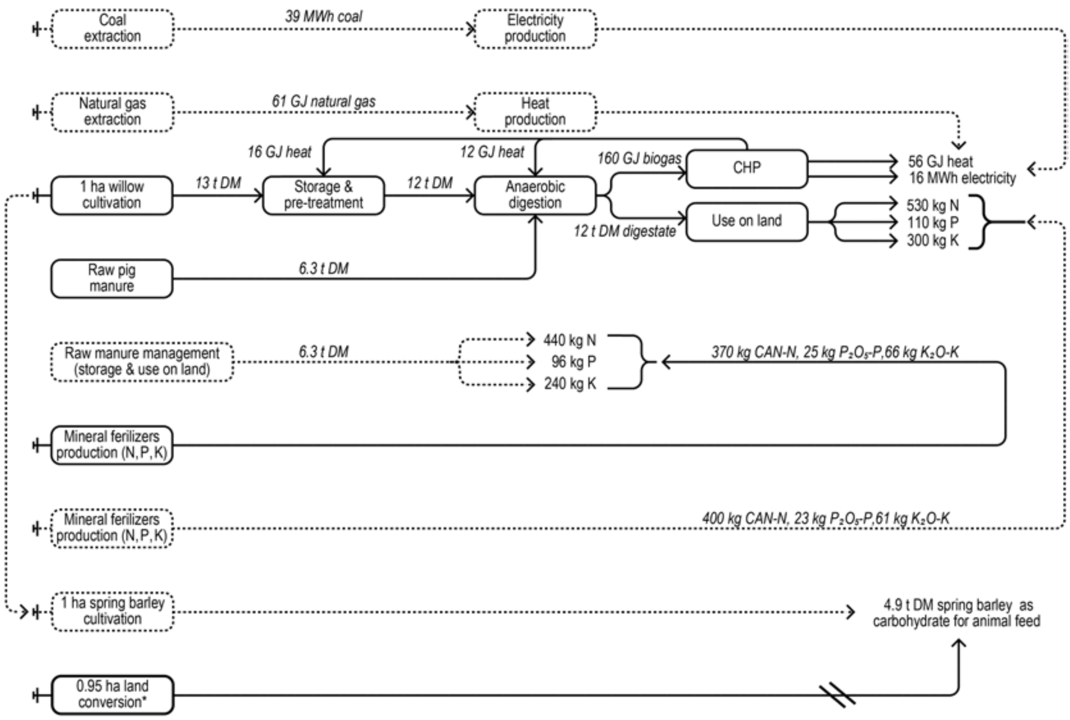

Figure S4. Process flow diagram for anaerobic co-digestion of willow with raw pig manure. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Values are rounded to 2 significant digits.

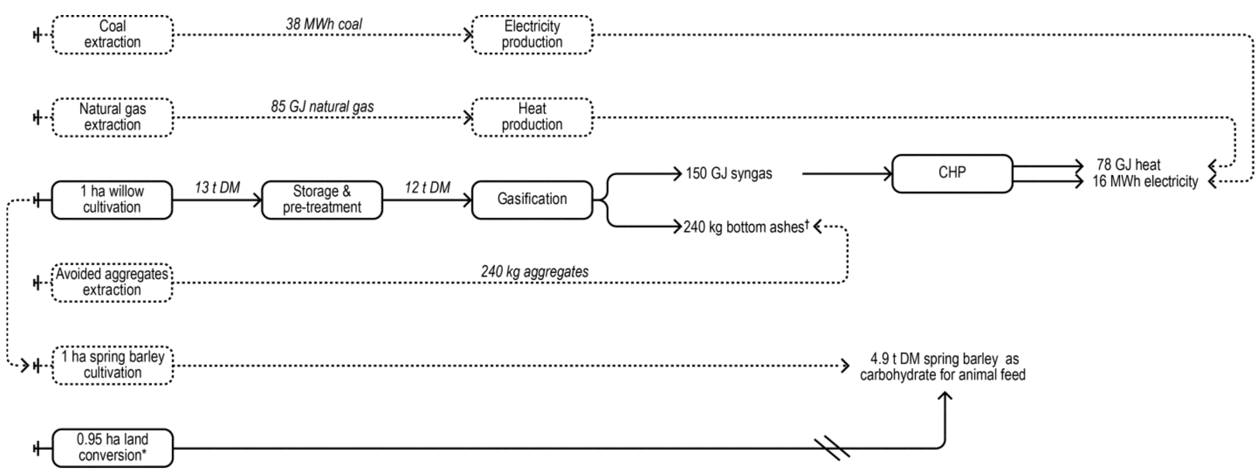

Figure S5. Process flow diagram for gasification of willow. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended). Values are rounded to 2 significant digits.

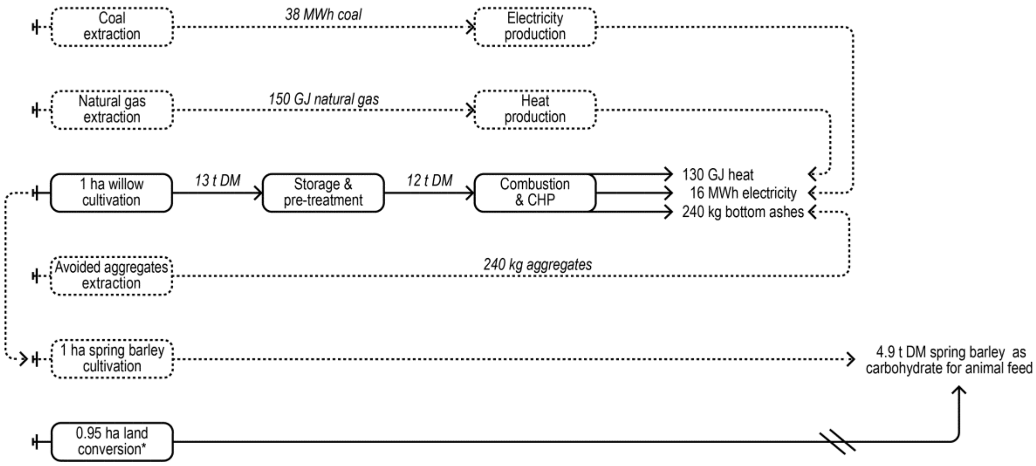

Figure S6. Process flow diagram for combustion of willow. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended).Values are rounded to 2 significant digits.

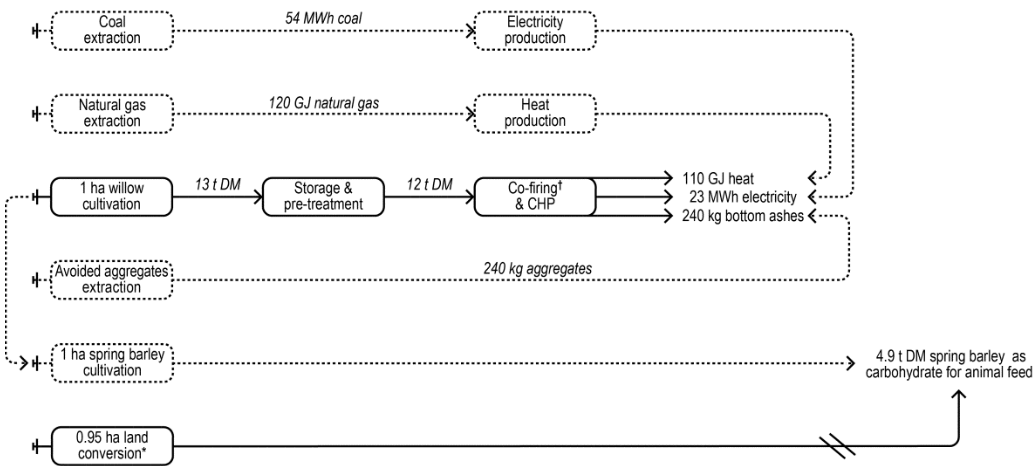

Figure S7. Process flow diagram for co-firing of willow. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Based on the data from co-firing Danish plants, the coal that is used here would have otherwise been used for CHP production, at similar conversion efficiency. Further, fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended). Values are rounded to 2 significant digits.

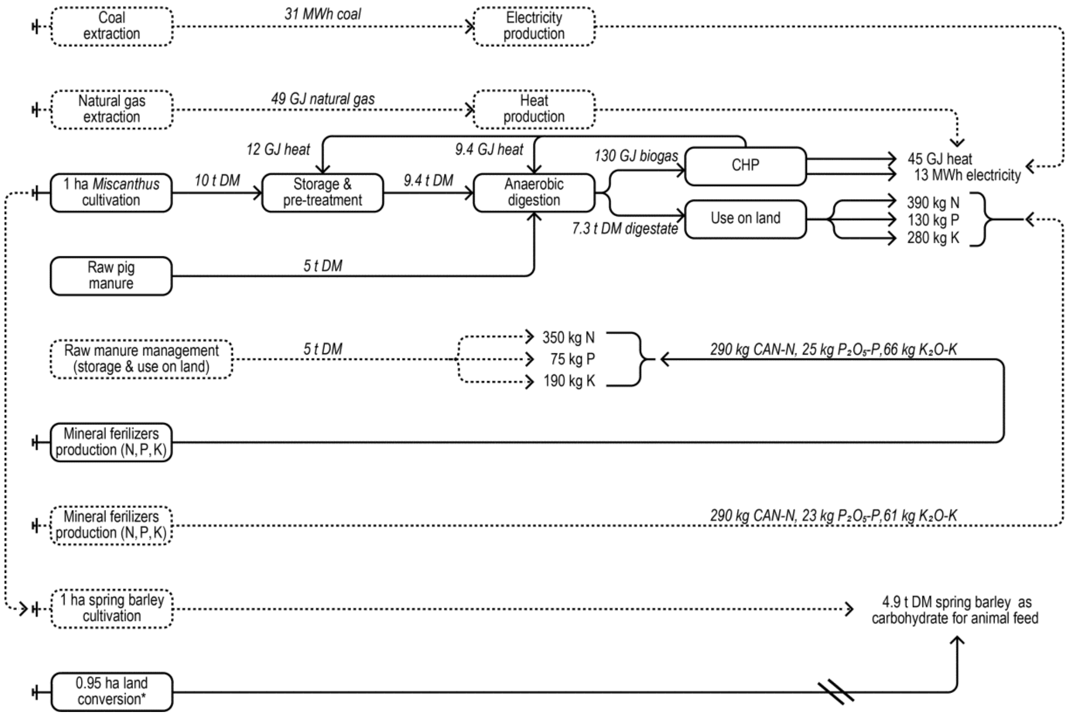

Figure S8. Process flow diagram for anaerobic co-digestion of *Miscanthus* with raw pig manure. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Values are rounded to 2 significant digits.

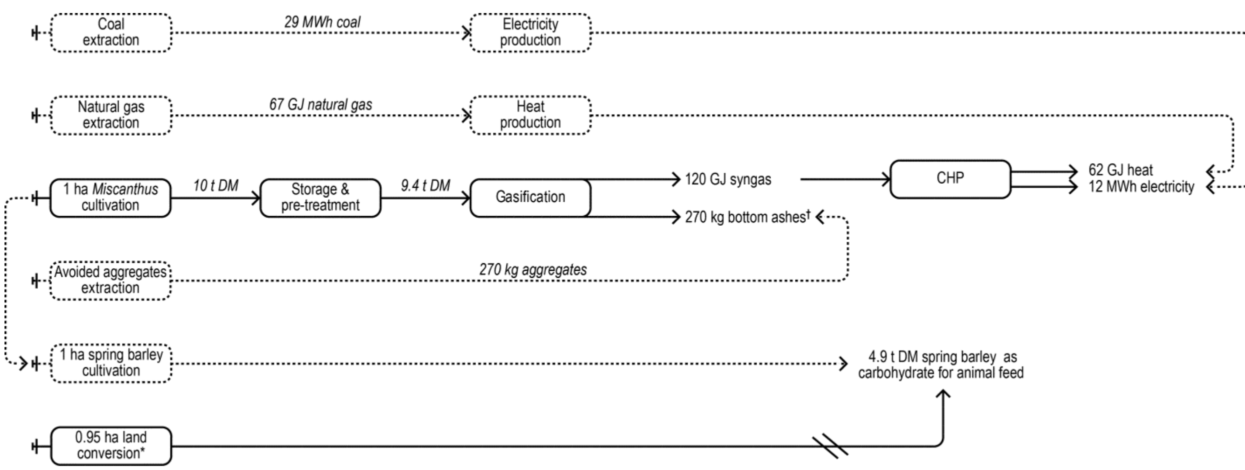

Figure S9. Process flow diagram for gasification of *Miscanthus*. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended). Values are rounded to 2 significant digits.

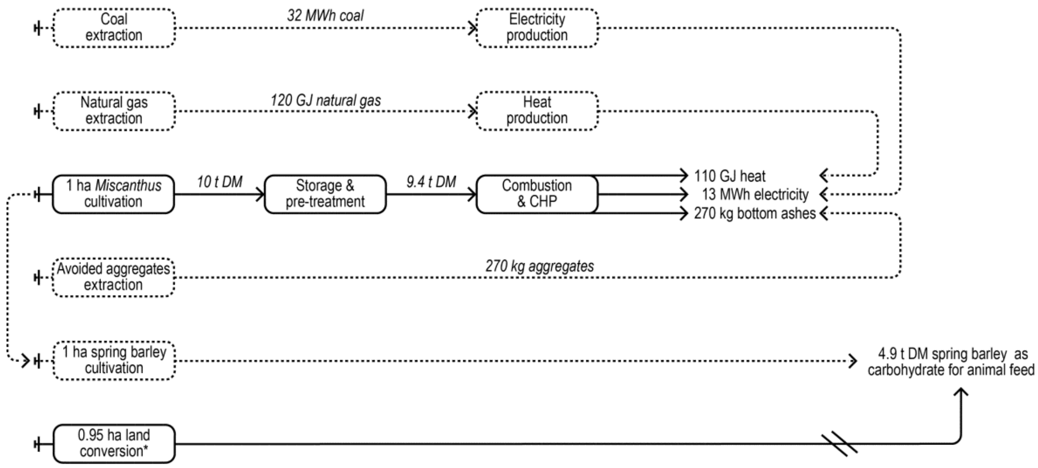

Figure S10. Process flow diagram for combustion of *Miscanthus*. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Values are rounded to 2 significant digits.

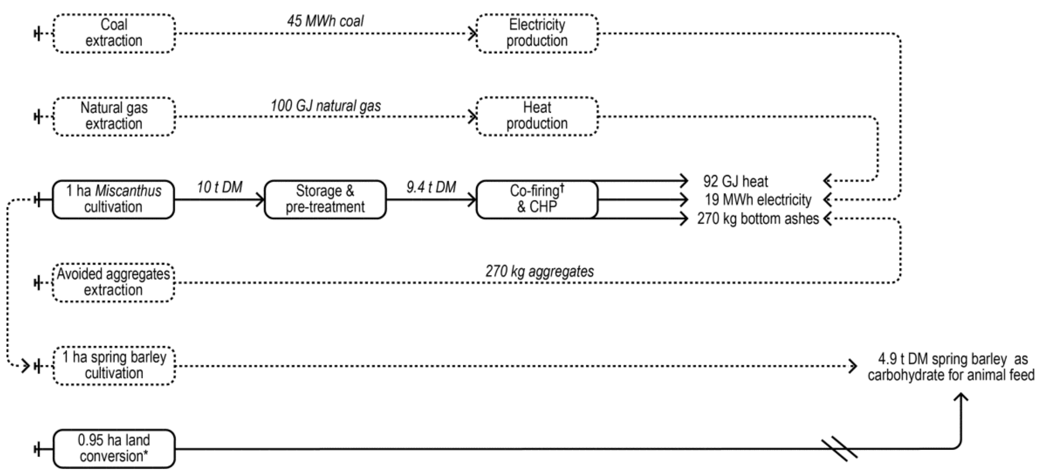

Figure S11. Process flow diagram for co-firing of *Miscanthus.* (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Based on the data from co-firing Danish plants, the coal that is used here would have otherwise been used for CHP production, at similar conversion efficiency. Further, fly ashes are considered used as back-filling material in old salt mines, and the environmental impacts from this are considered negligible (therefore the system boundary is not further extended). Values are rounded to 2 significant digits.

2. Identification of Marginals

2.1 Marginal energy technologies

Special attention was devoted to assumptions regarding the surrounding energy system as choices here may significantly affect the outcome of the LCA (1-6).

The purpose of bioenergy production is the decommissioning of fossil based energy production capacities (both electricity and heat) as these technologies are generally intended to be phased out in order to comply with political CO_2 reduction targets. Under this condition the electricity and heat produced from the selected bioenergy scenarios were assumed to substitute for the respective marginal fossil sources. The bioenergy scenarios were therefore credited with the environmental savings induced by substitution of fossil fuel-based energy production; such system boundary expansion to include the benefits deriving from replacement of fossil energy represents a typical approach in consequential LCA (e.g., (3-5) among the others). Of the fossil fuels, coal and natural gas represent the two ends of the range with respect to CO_2 emissions per combustion unit of fuel energy. In the baseline of the LCA, substitution of electricity produced from coal-fired power plants was assumed. With respect to Danish conditions this choice is supported by a number of studies (5, 6). This assumption was tested in the sensitivity analysis by substituting electricity produced from natural gas-fired power plants.

As opposed to electricity, the market for heat is rather local and substitution of district heating or heating fuels often depends on local conditions and production capacities connected to the district heating network in question (1). This means that when evaluating a system in a short term perspective involving existing production capacities, substitution of district heating should reflect local conditions. However, it is viable to assume that in the long term (with increasing bioenergy production) heat production from biomass will contribute to phasing out fossil fuels. With regard to the Danish market for heat, natural gas was identified as the fuel which is most likely to react to a marginal change in the heat demand/supply market. This choice is supported by (7). This assumption was tested in the sensitivity analysis by substituting heat produced from coal-fired power plants.

2.2 Marginal fertilizers

As illustrated in Figure 1 (of the main manuscript) for ryegrass and in Figure S4 (for willow) and S8 (for *Miscanthus*), the digestate produced from anaerobic digestion was used as a fertilizer (for N, P and K), which avoided marginal mineral N, P and K fertilizers to be produced and used, based on the content of N, P and K of the digestate. The marginal N, P and K fertilizers considered were calcium ammonium nitrate, diammonium phosphate and potassium chloride, respectively, conformingly with (8, 9).

3. Life-cycle inventory (LCI)

3.1 Crops

3.1.1 Ryegrass

The life cycle considered for perennial ryegrass is two years, which is common practice in Danish agriculture; sowing here occurs every second year, but harvests take place annually. Ryegrass is harvested in summer, swath and baled. The DM content considered at harvest is 20.5% (Table S1). The ryegrass is dried on field (to 85% DM content), stored indoor and further transported to the energy plant. The chemical composition and properties of the (today) Danish ryegrass are summarized in Table S1. For the storage and pre-treatments see section 3.2 and 3.3 of this document.

3.1.2 Willow

A 21 years life cycle has been considered for willow cultivation (6 cuts; 3 years harvest cycle, but first harvest occurring after 4 years; 1 year establishment; 1 year preparation before planting). Willow is harvested in the vegetative rest period (in the period around November to February). The water content considered at harvest is 50% (Table S1). The willow is harvested as whole rods, stored indoor and dried (to 85% DM content) and further transported to the energy plant. The chemical composition and properties of the willow are summarized in Table S1. For the storage and pre-treatments see section 3.2 and 3.3 of this document.

3.1.3 *Miscanthus*

The life time considered for a *Miscanthus* plantation in this study is 20 years (18 cuts; 1 year establishment: 1 year preparation before planting). Two harvest seasons are typically distinguished for *Miscanthus*, i.e., autumn and spring. Autumn harvesting is characterized by higher yield and higher concentration of water, nutrients and alkali. Delaying the harvest to spring lead to obtain a crop with better physical and chemical properties for thermal utilization, e.g., lower water content (below 20%), lower alkali content (e.g., Cl, K, N, S) as well as decreased ash content (10, 11). On the other hand, a delayed harvest lead to a decreased dry matter yield (i.e., 10 t DM ha^{-1} y^{-1} instead of about 15 t DM ha^{-1} y^{-1}) conformingly with (9), due to the loss of leaves. In this study only spring harvesting was considered for the assessment of all BtE conversion pathways. The authors are aware that for the specific case of anaerobic digestion, autumn harvested *Miscanthus* might be prioritized over spring's for its higher yield; however, spring harvest was assumed for all bioenergy scenarios in order to have the same assumptions regarding direct land use changes and storage across the *Miscanthus* scenarios. Furthermore, i) the data on *Miscanthus* pre-treatment and methane production were based on spring harvest; ii) scarce information was available on losses and type of storage of autumn harvested *Miscanthus* and unpublished studies reviewed by the authors showed DM losses up to 30% which would make the autumn harvest quantitatively comparable to the spring's when considering the storage.

It is considered that spring harvested *Miscanthus* is mowed and baled by a big baler. The harvested *Miscanthus* is then stored indoor and further transported to the energy plant. Spring harvested *Miscanthus* bales can be whether shredded (gasification and co-firing) or used directly (combustion). The chemical composition and properties of the *Miscanthus* (spring) are summarized in Table S1. For the storage and pre-treatments see section 3.2 and 3.3 of this document.

Table S1. Selected properties of the perennial energy crops evaluated in this study. In brackets the uncertainty range corresponding to the 95% confidence interval (i.e., the interval of length equal to four times the standard deviation around the mean) is reported. LHV_{db}: lower heating value (dry basis); LHV_{wb}: lower heating value (wet basis); LHV_{ar}: lower heating value as received (i.e., energy of the crop as fed into the energy plant after pre-treatment); CH_4 pot: methane potential; n.a.: not available.

Parameter	unit	Ryegrass	Willow	*Miscanthus*
Yield	t DM ha^{-1}	13.6 (±4.5)	12.7 (±4)	10 (±3.3)
DM (at harvest)	% FM	20.5 (±1.7) †	50 (±5)$^{\gamma}$	90 (±6) $^{\beta}$
VS	% DM	92.3 (±1) †	98.1 (±1.8)*	n.a.
Ash	% DM	7.7 (±1) †	1.9 (±0.9)*	2.7‡
C	% DM	46.4 (±2.2)*	48.9 (±1)*	47.7 (±1)*
H	% DM	5.7 (±0.3)*	6.0 (±0.2)*	5.5 (±0.3)*
N	% DM	2.9 (±0.6) †	0.6 (±0.3)*	0.44 (±0.13) $^{\alpha}$
P	% DM	0.40 (±0.08) †	0.07*	0.49 (±0.08) $^{\alpha}$
K	% DM	0.33 (±0.06) †	0.3*	0.69 (±0.2) $^{\alpha}$
HHV	MJ kg^{-1} DM	18.0 (±2.5)*	19.4 (±0.8)*	19.0 (±0.6)*
LHV_{db}	MJ kg^{-1} DM	16.8 (±2.4)*	18.1 (±0.8)*	17.8 (±0.6)*
LHV_{wb}	MJ kg^{-1} DM	1.5 (±1)	7.9 (±0.6)	16 (±0.5)
LHV_{ar}	MJ kg^{-1} FM	14 (±2)	15 (±0.6)	16 (±0.5)
CH_4 pot	$Nm^3 CH_4$ t^{-1} VS	410$^{\varepsilon}$	350$^{\varepsilon}$	360$^{\varepsilon}$

* Based on (12).
† Based on (13). After on field drying, the DM content is assumed 85% FM.
‡ Based on (14).
α Based on (15).
β Based on (9, 16)
γ Based on (17).
ε See section 3.4.1.

3.2. Crop storage

Storage is needed within the bioenergy chain as biomasses accumulate seasonally and the energy plants have, instead, to be fed and run continuously. Furthermore, biomass prices will be market-driven and the producers will sell the crops whenever the prices will be convenient, therefore storage will be applied. Storage conditions have been modeled according to available literature on biomass dedicated to energy and feed; the main environmental issue of the storage is the dry matter losses which cause (primarily) a decrease of the available biomass and emissions of CO_2, CH_4, NH_3, and N_2O due to carbon and nitrogen degradation.

For dry herbaceous species, i.e., ryegrass (after on field drying to achieve DM content of 85%) and spring harvested *Miscanthus* (DM 90%) dry matter losses of 5.5% (±4.5%) were considered based on (18). These values are in accordance with other studies focusing on grass storage for feed production (19, 20). For willow, the storage was modeled as 'whole rods storage' which also represents a method typically applied to dry the harvested willow stems over summer (17, 21-27); this way, the storage also functions as a drying pre-treatment. This choice of storage condition was supported by the fact that other conditions were less beneficial, e.g., storage of wet willow chips was proved to determine higher dry matter losses as a consequence of increased microbial activity and degradation (17, 26, 28) and thermal drying is associated with significant economical and energy costs which make it less attractive (29). The dry matter losses reported by a number of experimental studies (17, 21, 26) for storage-drying of willow rods ranged between 3.5%-6.1% (average value assumed 4.8%).

In this study only the emissions of CH_4 and CO_2 caused by dry matter degradation and losses within the storage period were modeled based on the assumed dry matter losses. The CO_2 emission was calculated proportional to the total dry matter loss based on the concentration of carbon in the dry matter, assuming aerobic conditions. The CH_4 emissions associated with crop and digestate storage were estimated based on the tier 2 IPCC approach for manure management (30), considering a methane conversion factor (MCF) of 0.5% and 1%, respectively. The emissions of N_2O, NH_3 and NO_3 (to surface water) were not included as the research on these is still at an early stage (18). However, the overall nitrogen mass losses were estimated based on the C to N ratio (i.e., the loss of nitrogen was proportional to the carbon loss based on the ratio

C/N in the crop). The C and N losses are shown in Figure S13-S18. Indoor storage of the crops was assumed (duration longer than 4 months).

The authors are aware of that other storage techniques exist, e.g., ensiling for anaerobic digestion. However, dry storage was assumed for all bioenergy scenarios in order to have consistency regarding storage assumptions across the assessed bioenergy scenarios. Furthermore, with respect to co-digestion crop-manure, the energy production per unit-input increases with the dry matter content of the co-substrate (i.e., crop) (see 3.4.1). Therefore, if the idea is to use the crops for boosting manure digestion dry biomass will be preferred over wet substrates. Furthermore, handling and storage of dry biomass is easier and associated with less dry matter losses and emissions (18). The influence of the variation of the parameters used to model the storage on the final LCA results has been assessed in the uncertainty analysis.

3.3 Pre-treatments

An overview is presented in Table S2. Follows a detailed description of the pre-treatments modelled in the LCI.

3.3.1 Pre-treatments: anaerobic digestion

Ryegrass has a particularly high water content (ca. 80% of FM) at harvest. Therefore, a drying process is needed for ryegrass when undergoing a thermal energy conversion. On field drying was thus considered and modelled based on the on field drying process traditionally used for hay: the ryegrass is left on field for a few weeks, where it is mowed and turned in order to facilitate the drying to DM content of 85%. The dry matter losses caused by microbial respiration as well as by the different operations was estimated to 20% (±10%) of the initial DM content (20, 31). Biomasses undergoing anaerobic digestion require size comminution (assumed 10-50 mm); this was considered by including an electricity consumption of 7.5 kWh t^{-1} DM (32). Given their high lignin content, *Miscanthus* and willow are rather resistant to microbial degradation. A pre-treatment is therefore necessary in order to break the lignocellulosic structures of these energy crops and render a maximum of their C content bioavailable. For both crops, a thermal treatment has thus been considered (33, 34), and this was accounted for in the LCA as decreased heat production (hence decreased substitution of heat from natural gas). Based on (33), the heat required for the pre-treatment corresponded to about 1.3 GJ t^{-1} DM.

3.3.2 Pre-treatments: gasification

The gasification process in fluidized bed typically requires biomass with water content below 20% (35). For ryegrass the on field drying was assumed as described in 3.3.1. Before energy conversion, size comminution (10-50 mm) was assumed, as for anaerobic digestion. This way, the biomass bales (i.e., ryegrass and *Miscanthus*) or rods (i.e., willow) are loosened/comminuted and homogeneous process conditions are facilitated. For willow, natural drying (down to water content of 15%) was assumed to occur during indoor storage of 'whole rods' based on experimental studies (17, 21, 26). Size comminution (10-50 mm) was assumed before energy conversion, as for ryegrass. No drying was taken into account for the gasification of spring harvested *Miscanthu*s, due to its low water content (10%) at harvest. Size comminution (10-50 mm) was assumed required as for ryegrass and willow.

3.3.3 Pre-treatments: combustion and co-firing

For combustion and co-firing the approach for the drying process was the same as for gasification. For combustion in small-to-medium scale biomass CHP plants no other pre-treatment was included. In fact, ryegrass and *Miscanthus* bales as well as willow 'whole rods' can be fed directly when combusted in these plants which have been optimized in the last decades to burn locally available biomasses without the need for expensive pre-treatments such as pelletization, shredding and pulverization. On the other hand, the electricity recovery decreases as a consequence of the lower plant size and fuel quality. Nevertheless, small-to-medium scale biomass CHP plants have been optimized (in some cases with flue gas condensation) to recover as much as 90% of the initial energy of the fuel in form of heat for district heating purposes. This is already done for biomasses with similar characteristics to *Miscanthus* and willow such as straw and wood chips. For co-firing instead, size comminution (10-50 mm) was included in the model. Pelletization and milling of the pellets were not included in the baseline calculation (as parallel co-firing was assumed). However, this assumption was tested in the sensitivity analysis by including pelletization and milling prior to direct co-firing.

Table S2. Overview of pre-treatments and energy efficiency of the BtE conversion technologies considered in this study (rounded values). In brackets the uncertainty range corresponding to the 95% confidence interval (i.e., the interval of length equal to four times the standard deviation around the mean) is reported. AD: anaerobic co-digestion of energy crops with raw pig manure; GA: thermal gasification; CO: direct biomass combustion in small-to-medium scale CHP plants; CF: co-firing in large scale coal-fired CHP plants.

		Pre-treatment					Energy conversion			
BtE		Steam	Drying	Comminution	Pelletizing	Milling	BtE Technology	η_{el} (%)	η_{th} (%)	η_{tot} (%)
Ryegrass	AD		X	X			Gas engine	38 (±4)	52 (±8)	90 (±5)
	GA		X	X			Gas engine	38 (±4)	52 (±8)	90 (±5)
	CO		X				Steam cycle	27 (±2)	63 (±7)	90 (±5)
	CF		X	X	X*	X*	Steam cycle	38 (±3)	52 (±8)	90 (±5)
Willow	AD	X		X			Gas engine	38 (±4)	52 (±8)	90 (±5)
	GA		X	X			Gas engine	38 (±4)	52 (±8)	90 (±5)
	CO						Steam cycle	27 (±2)	63 (±7)	90 (±5)
	CF		X	X	X*	X*	Steam cycle	38 (±3)	52 (±8)	90 (±5)
Miscanthus	AD	X		X			Gas engine	38 (±4)	52 (±8)	90 (±5)
	GA			X			Gas engine	38 (±4)	52 (±8)	90 (±5)
	CO						Steam cycle	27 (±2)	63 (±7)	90 (±5)
	CF			X	X*	X*	Steam cycle	38 (±3)	52 (±8)	90 (±5)

* Pelletizing and milling may be required when applying direct biomass co-firing with pulverized coal. This scenario was included in the sensitivity analysis only.

3.4 Biomass-to-Energy conversion technologies

3.4.1 Anaerobic digestion

Digestion of carbohydrate-rich energy crops alone (e.g., willow and *Miscanthus*) has the primary advantage of requiring significantly low digestion volumes because of the high dry matter content of the feedstock; this makes anaerobic digestion of such crops economically attractive. However, mono-digestion of energy crops may encounter a number of technical problems (and eventually failures) related to the sub-optimal content of micro-nutrients (e.g., nickel, cobalt, etc.) and macro-nutrients (high C to N ratio); recent studies have indicated the optimal C to N ratio to be between 16-20 (36-38) and have demonstrated how a sub-optimal concentration of selected micro-nutrients may lead to process failure (36). Co-digestion with nutrients-rich substrates such as the organic fraction of municipal solid waste or manure may solve these problems (36, 37). In addition, manure represents one of the most abundant domestically available biomass resources for Denmark (about 23-34 PJ) which is only to a minor extent (6% of the potential) exploited for energy production (39). The scarce economical and technical attractiveness of manure mono-digestion is primarily due to the low energy production per unit of reactor volume as a consequence of the extremely low dry matter content of the feedstock (between 2% and 10% depending of the type of animal manure). The current management in Denmark is by far (94% of the potential) represented by spreading on land of raw manure. This practice leads today to large environmental impacts on most environmental compartments, mainly global warming and eutrophication (8). Hence, co-digestion of manure and energy crops may represent a viable alternative to produce bioenergy and improve manure management.

In this study, a generic two-stage mesophilic anaerobic digestion plant was modeled, where the energy crops were co-digested with manure. The principal parameters modeled were: i) methane potential and yield, ii) ratio manure:crop in the mix fed, and iii) energy consumption for plant operation.

Based on (8), the i) methane potential of raw pig manure was 450 $Nm^3\ t^{-1}$ VS. The yield was set to 70%, i.e., generating 320 $Nm^3\ t^{-1}$ VS (8). The methane potential of the crops was calculated from the Buswell's equation based on the content of lipids, carbohydrates, proteins and lignin (Table S3). The methane potential was calculated to 410, 350 and 360 $Nm^3\ CH_4\ t^{-1}$ VS for ryegrass, willow and *Miscanthus*, respectively. The methane potential for willow and *Miscanthus* was calculated from the composition of the crops as after steam pre-treatment based on the results of (33). The pre-treatment determined a partial decomposition of the lignin structure so that more sugars were bioavailable for microbial degradation. This explained the difference between pre-treated and raw substrates. For all crops (as for manure) the methane yield in the digester (including post-digestion tank) was set to 70% of the methane potential based on literature (40). The corresponding methane production was therefore 290, 240 and 250 Nm^3 CH_4 t^{-1} VS. It was assumed that 90% of the total production occurred in the first digestion stage. Notice that the estimated methane yield for ryegrass was consistent with the values found in literature (198-510 Nm^3 CH_4 t^{-1} VS, see Table S4).

Table S3. Composition of ryegrass, willow and *Miscanthus* in terms of lipids, carbohydrates, proteins, lignin and relative calculated methane potential. $CH_{4\ pot}$: methane potential (Nm^3 t^{-1} VS); %VS (concentration of the parameter (e.g., lipids) as % of VS); D_{raw}: degradability of the raw substrate (% $CH_{4\ pot}$); D_{PT}: degradability of the pre-treated substrate (% $CH_{4\ pot}$). The values of D_{PT} for willow and *Miscanthus* are based on laboratory batch-tests (33).

Parameter	$CH_{4\ pot}$	Ryegrass			Willow			*Miscanthus*		
		%VS	D_{raw}	D_{PT}	%VS	D_{raw}	D_{PT}	%VS	D_{raw}	D_{PT}
Lipids	1014	4.3	100	-	0.0	-	-	0.0	-	-
Cellulose	415	47.6	100	-	41.2	60	100	47.6	60	100
Hemicellulose	415	15.5	100	-	14.9	70	100	18.5	70	100
Proteins	496	20.2	100	-	0.0	-	-	0.0	-	-
Lignin	200†	10.4	0	-	31.6	0	100	25.2	0	100
Residue	415	0.0	100	-	12.2	0	100	8.8	0	100
CO_2/CH_4		2.6			2.7			2.8		
$CH_{4\ pot}$ (crop)		410			350			360		

† Based on (33).

In order to ii) calculate the ratio manure:crop (and so the amount of manure utilized and digested per hectare of the crop-system) a mass balance based on (8) was established (Eq. S1-S6). This allowed calculating the ratio manure:crop for different values of dry matter of the digestate obtained after the first digestion stage. A DM content of 10% in the digestate represents an upper constraint in order to assure the pumpability of the digestate in wet digestion systems (8). This constraint determines the maximum amount of co-substrate (e.g., crop) that could be mixed with the manure.

$$DM_{digest} = \frac{\lfloor (W_{man} \cdot DM_{man}) - VS_{deg,man} \rfloor + \lfloor (W_{crop} \cdot DM_{crop}) - VS_{deg,crop} \rfloor}{(W_{man} - W_{biogas,man}) + (W_{crop} - W_{biogas,crop})} \quad \text{Eq S1.}$$

$$1000 = W_{man} + W_{crop} \quad \text{Eq S2.}$$

$$W_{biogas,man} = VS_{man} \cdot W_{man} \cdot yield_{man} \cdot \rho/(CH_4\% \cdot 1000) \quad \text{Eq S3.}$$

$$W_{biogas,crop} = VS_{man} \cdot W_{man} \cdot yield_{crop} \cdot \rho/(CH_4\% \cdot 1000) \quad \text{Eq S4.}$$

$$VS_{deg,man} = W_{man} \cdot DM_{man} \cdot VS_{man}/DM_{man} \cdot DR_{man} \quad \text{Eq S5.}$$

$$VS_{deg,crop} = W_{crop} \cdot DM_{crop} \cdot VS_{crop}/DM_{crop} \cdot DR_{crop} \quad \text{Eq S6.}$$

Where:

DM_{digest}: DM of the digestate after the first digestion stage (% FM)
W_{man}: weight of the manure input (kg)
DM_{man}: DM of the manure input (% FM)
$VS_{deg,man}$: VS degraded from the raw manure after the first digestion stage (kg)
W_{crop}: weight of the crop input (kg)
DM_{crop}: DM of the crop input (% FM)
$VS_{deg,crop}$: VS degraded from the crop after the first digestion stage (kg)
$Yield_{man}$: methane yield of the manure after the first digestion stage ($Nm^3\ t^{-1}$ VS)
$Yield_{crop}$: methane yield of the crop after the first digestion stage ($Nm^3\ t^{-1}$ VS)
ρ: biogas density (kg Nm^{-3})
DR_{man}: degradation rate of the manure after the first digestion stage (% VS)
DR_{crop}: degradation rate of the crop after the first digestion stage (% VS)

The DM (as % FM) and VS (as % DM) content of the manure was 6.97% and 80%, based on (8). The DM content of the crops ex-storage was assumed equaled to the DM content prior to storage. The degradation rate of the manure after the first stage was 60% based on (8). For the crops, it was calculated based on Eq. S7, and equaled 54%, 46% and 47% for ryegrass, willow and *Miscanthus*, respectively. The yield of methane in the first digestion step was assumed 90% of the total. The remaining 10% was assumed produced and collected in the post-digestion tank. The density ρ of the biogas was 1.158 kg Nm^{-3} based on CH_4 content in the biogas of 65%.

$$\frac{DR_{man}}{CH_{4\,pot\,man} \cdot Tot\ Yield_{man}} = \frac{DR_{crop}}{CH_{4\,pot\,crop} \cdot Tot\ Yield_{crop}} \quad \text{Eq. S7.}$$

Where:

DR_{man}: degradation rate of the manure after the first digestion stage (% VS)

DR_{crop}: degradation rate of the crop after the first digestion stage (% VS)

$CH_{4\ pot\ man}$: methane potential of the manure ($Nm^3\ CH_4\ t^{-1}$ VS)

$CH_{4\ pot\ crop}$: methane potential of the crop ($Nm^3\ CH_4\ t^{-1}$ VS)

Tot $Yield_{man}$: total methane yield of the manure (% $CH_{4\ pot\ man}$)

Tot $Yield_{crop}$: total methane yield of the crop (% $CH_{4\ pot\ crop}$)

The results are presented in Figure S12 with respect to different dry matter of the digestate (5% to 10%) obtained after the first digestion stage. It is evident that the biogas plant operators will utilize as much crop as possible to boost the energy production per unit of feed input. The energy production will be maximized for a digestate at DM equal to 10% corresponding to a ratio (fresh matter basis) manure:crop of 5.7, 6.4 and 6.7 for ryegrass, willow and *Miscanthus*, respectively. The amount of manure utilized for co-digestion was therefore 69, 92 and 72 t FM ha^{-1} for ryegrass, willow and *Miscanthus*, respectively.

With respect to iii) electricity and heat consumption for the plant operation the data were based on (8): the electricity consumption was set to 2% of the overall energy in the produced biogas (corresponding to about 5% of the net electricity production) and the heat consumption was calculated based on the thermal energy required to heat up manure and crops from 8 ºC to 37 ºC. The fugitive emission of methane was estimated to 1% of the methane produced, based on recent LCA studies (8, 32, 41). Emissions of biogenic CO_2 were estimated as a function of the biogenic CH_4 releases, based on the methodology described by (8). Based on this, the ratio CO_2 to CH_4 was found to correspond to 2.6 for ryegrass, 2.7 for willow and 2.8 for *Miscanthus* (Table S3).

The biogas generated from anaerobic digestion was assumed to be used in a gas engine with an average electricity efficiency of 38% (±4%), based on a review of different gas engine technologies (42). The total energy efficiency was set to 90%, thus raising heat recovery efficiency to 52% (Table S2). The total energy efficiency was based on a review of a number of small-to-medium scale biomass CHP plants (section 3.4.3). Similar values are reported by (43). The emissions associated with the biogas combustion in gas engines were based on (44) (Table S7). The environmental savings and impacts associated with the management (i.e., storage, digestion and use on land) of the manure were accounted for based on previous results (8) (the LCA system boundary was therefore expanded accounting for the amount of manure utilized and digested in each crop-system).

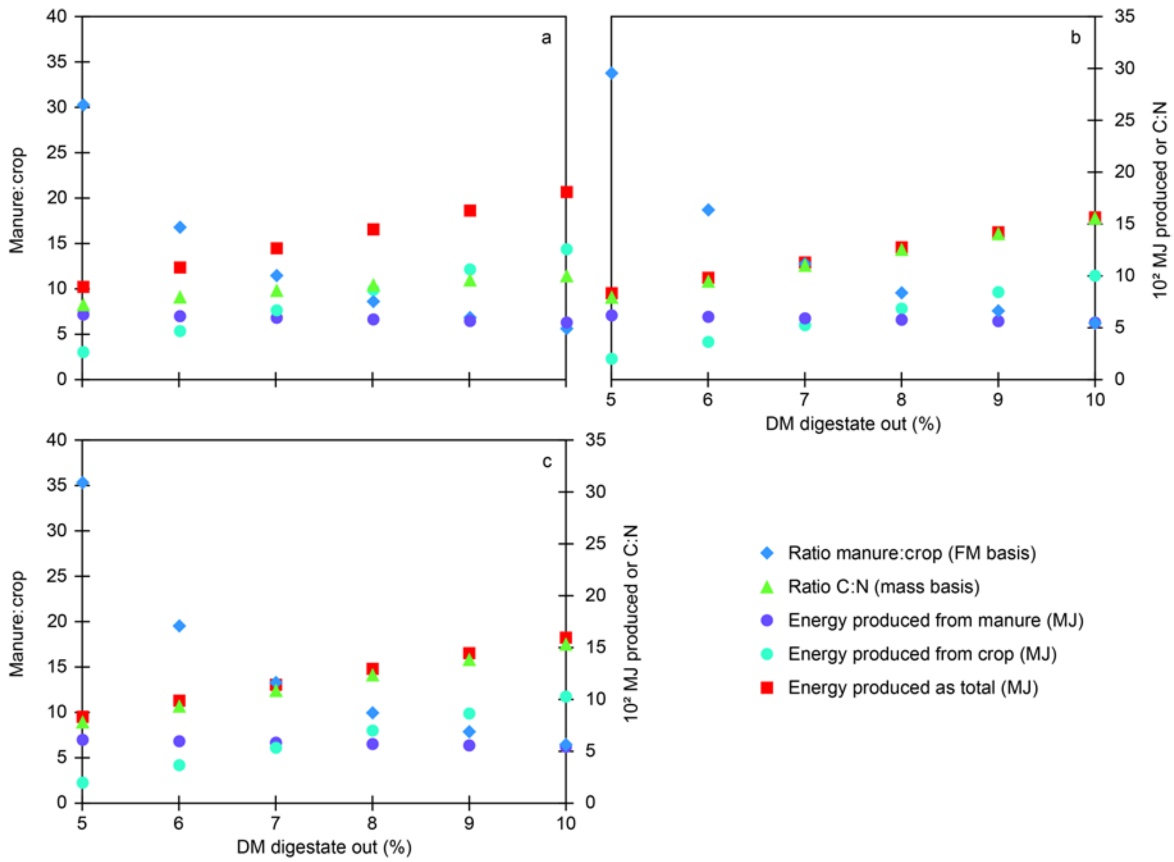

Figure S12. Illustration of I) share of manure (kg t^{-1} FM input) in the mix manure-crop fed into the digestion plant, II) C to N ratio of the mix manure-crop fed into the digestion plant, III) share of the total energy produced from manure (MJ t^{-1} FM input), IV) share of the total energy produced from the crop (MJ t^{-1} FM input) and V) total energy produced (MJ t^{-1} FM input) as a function of the dry matter content of the digestate obtained after the first digestion stage; a) ryegrass; b) willow; c) *Miscanthus*.

Table S4. Overview of methane yield (or potential) reported in the reviewed literature studies.

Biomass	CH_4 yield ($Nm^3 CH_4\ t^{-1}$ VS)	Note	Source	This study
Ryegrass	198-360	Lab batch test 38 ºC, 35-40 days	(45)*	290
	233-327	Lab batch test 35 ºC, 28 days	(46)*	
	300-320	Lab/semi-continuous/35 ºC/28 days	(47)*	
	320-510†	Lab batch tests/70-80 days	(48)*	
	310-360	Lab batch tests/35 ºC/28 days	(49)*	
	410†	Lab and pilot scale tests	(50)	
	390	Pilot scale	(51)	
	361†	Lab batch test	(52)	
Other grasses	197-470	Review of different grass species	(53)	-
	305	Modeling	(54)	
Willow	300	With pretreatment	(33)	240
	90	No pretreatment	(51)	
Miscanthus	300	With pretreatment	(33)	250

*Tabulated in (31).
† Methane potential.

3.4.2 Gasification

A generic fluidized bed reactor was modeled based on existing pilot plants (32, 55-57). The main parameters modeled were: cold gas and carbon conversion efficiency (CGE and CCE), energy content of the syngas and energy consumption of the plants. The CGE defines the fraction of the feedstock chemical energy (as LHV, dry basis) remaining in the syngas (and not lost as, e.g., heat or in the residue). It is expressed as the ratio between the amount of energy in the syngas (after gas cleaning) and the amount of energy in the biomass (as LHV, dry basis). The CCE defines the proportion of the feedstock C that is transferred to the syngas (as CH_4, CO and CO_2 and then to CO_2 after further syngas combustion).

The data for CGE and CCE were based on a number of literature studies focusing on woody and herbaceous biomass (Table S5). In general, the energy conversion efficiency for high quality woody biomass (e.g., high quality wood pellets from forest trees) was higher than for low grade wood (e.g., waste wood), fast-growing trees (e.g., willow) and herbaceous crops (e.g., grasses and *Miscanthus*). The energy conversion efficiency for herbaceous biomass (e.g., grass and *Miscanthus*), willow and waste wood was the lowest. However, other studies based on modeling of gasification processes (58, 59) indicated higher efficiencies (about 85%) for gasification of lignocellulosic and herbaceous energy crops. The difference between modeling and pilot-scale experimental results is associated with the high heat losses typically occurring in small-scale pilot plants (up to 10-20%); this is often the reason why these facilities do not reach high CGE efficiencies. Therefore, based on the data reported in Table S5, the CGE (for all 3 crops) was equaled to the mid value of the large range 55%-85%. (i.e., 70% ±15%). Also from the above-mentioned literature review, the CCE was equaled to the mid value of the range 91-99% (i.e., 95% ±4%). The influence of the variability of both the CGE and the CCE on the final LCA results was assessed in the uncertainty analysis.

The consumption of electricity to operate the plant ranged between 26 (without biomass comminution) and 30 (with biomass comminution) kWh t^{-1} DM (32). The syngas was assumed to be used in a gas engine yielding the same efficiency as when burning the biogas (Table S2). The consumption of bed materials and chemicals to run the plant was based on (32). The emissions associated with the combustion of syngas in gas engines (Table S7) were based on (44).

Table S5. Overview of CGE and CCE reported in the reviewed studies on gasification of different woody and herbaceous biomasses; CFB: circulating fluidized bed; FB: fluidized bed; BFB: bubbling fluidized bed; n.a.: not available.

Biomass	H_2O (% FM); ash (% DM)	CGE (%)	CCE (%)	Technology	Source
Grass pellets (verge)	7.3; 17.6	58-64	92.7-94.7	CFB; air gasification	(57)
Grass pellets (switchgrass)	8.38; 8	62	n.a.	FB; steam gasification	(60)
Miscanthus pellets	6.78; 1.2	73	n.a.	Fixed bed; steam gasification (Lab -scale)	(61)
Willow chips	17; 2.1	66	91.7-97	CFB; air gasification	(57)
Willow pellets	8; 2.52	55.2-62	86.9-92	CFB; steam-O_2 blown gasification	(62)
Wood pellets (larch)	8.16; 0.12	79.6	96.9	BFB; air gasification	(63)
Wood pellets (cedar)	10; 0.3	82	99	FB; air gasification	(64, 65)
Wood pellets	6.3; 0.5	68	97	BFB; air-steam gasification	(66)
Wood pellets	4.56; 0.4	60	n.a.	FB; steam gasification	(60)
Wood pellets	6.7; 1	70-84	n.a.	BFB; air gasification	(67)
Wood pellets	8; 0.3	69	92	BFB; air gasification	(68)
Waste wood	16; 8	62-70	87-99	BFB; air gasification	(68)
Waste wood	7-11; 1.2-3.3	49-66	n.a.	BFB; air gasification (mainly)	(67)
Wood chips (oak, beech)	32.2; 0.9	93	99.4	Two-stage gasifier with pre-heating and pyrolisis of the wood chips	(69)
Grass, straw, wood	n.a.	80-85	n.a.	Modeling	(58)
Grass	n.a.	79.8	n.a.	Modeling	(59)

3.4.3 Combustion and co-firing with fossil fuel

For direct biomass combustion, a generic small-to-medium scale (1-100 MW of net power output) biomass CHP plant was modeled based on a number of reviewed centralized and decentralized biomass CHP plants established essentially in Denmark (Table S6). The decentralized biomass CHP plants ranged from small-to-medium scale (1-100 MW of net power output in full-load). The centralized (large scale) CHP power plants were *Avedoerevaerket* and *Oestkraft*; in particular, *Avedoerevaerket* is considered as one of the most efficient existing co-firing CHP plants; the net power output in full-load is 355 MW (without gas turbine) – 495 MW (with gas turbine) in CHP mode and 425 MW (without gas turbine) - 575 MW (with gas turbine) in condensing mode. The reported efficiencies (Table S6) refer to the net full-load electricity and heat efficiency (i.e., own plant consumption for biomass handling, shredding, milling etc. has been subtracted), if not otherwise specified. In the LCA model, the direct combustion of ryegrass/*Miscanthus* (bales) and willow (chips) was modeled similarly to, respectively, straw and wood chips combustion (for the following processes: handling, feeding and air emissions). This is supported by the fact that the composition as well as the water content of herbaceous biomass and willow chips is similar to straw and wood chips, respectively, and by the fact that previous tests realized in Danish power plants have shown similar combustion efficiencies and behaviors (11, 70). Secondly, it is envisioned to be likely that biomass producers and energy operators will use established harvesting/baling machines (already in use for straw and wood chips) as well as power plant technologies (already developed for straw and wood chips) for handling the “new” biomasses with as little as possible technical adaptations, thus avoiding expensive investments in new technologies. The net electricity efficiency (full-load) in the reviewed biomass plants ranged from 13% (for old plants and plants co-firing waste and natural biomass) to 29% (best available technologies such as *Maribo-Sakskoebing* and *Herning*). The total efficiency (full-load) ranged from 76% to 96%. However, for the recently commissioned plants and the installations combusting only biomass (e.g., straw and wood chips), the electricity efficiency was typically found in the range 25% (*Assens*)-29% (*Maribo/Sakskoebing*). In this study the net electricity efficiency was therefore assumed equal to the mid value of this range (27% ±2%). The associated total efficiency was between 85% and 95% with average 90% (±5%). In the uncertainty analysis the influence of the variation of the energy efficiency on the LCA results was assessed. At this stage of the research, the information and the literature regarding the air emissions (other than CO_2) from combustion of ryegrass, willow and *Miscanthus* in biomass CHP plants is scarce. Therefore, based on the chemical composition, the air emissions from combustion of straw (44) were used as proxy for ryegrass and *Miscanthus*, whereas the air emissions from wood chips (44) were used as proxy for willow (Table S7). The consumption of resources and material to operate the plant was based on (71).

With respect to co-firing of the biomasses with fossil fuel, three main configurations exist: direct co-firing (the biomass, typically as pellets, is milled/pulverized together with coal and fired in the same system), indirect co-firing (the biomass is gasified and then the syngas is fired along with fossil fuel in the same system) and parallel co-firing (the biomass is combusted in separate boiler; the steam generated is used in the same steam turbine as for the steam derived from fossil fuel combustion, with high efficiency). An example of world-wide best available technology for parallel as well as direct co-firing is *Avedoerevaerket* power plant where parallel co-firing of straw and direct co-firing of wood pellets (milled/pulverized and fired along with coal) is operated. Wood chips can also be used as fuel for parallel co-firing. This is demonstrated by the fact that in periods where straw was not harvested (too humid because of wet summers), wood chips were used instead. With respect to crops storage, handling and feeding, the co-firing technology was modeled based on this specific power plant. The (full-load) electricity efficiency of the reviewed co-firing CHP plants was in the range 35% (*Oestkraft*) - 41% (*Avedoerevaerket*). The mean value (38%) was assumed for the baseline modeling (uncertainty ±3%). This was also the average annual net electricity efficiency of *Avedoerevaerket*. The related total efficiency was set to 90% (±5%) as for direct biomass combustion. In the uncertainty analysis the influence of the variation of the energy efficiency on the LCA results was assessed. The consumption of resources and material to run the plant was modeled based on (72). The air emissions were assumed the same as for direct biomass combustion.

Table S6. Overview of the (full-load) energy efficiencies of the reviewed biomass CHP plants. CP: condensing plant; CHP: combined heat and power plant; η_{el}: electricity efficiency; η_{tot}: total efficiency (heat plus electricity).

Type	Name	Fuel	Technology	η_{el}	η_{tot}
CP	*NEPCO* plant (-)	Wood (unspecified)	Travelling grate	29	-
CP	*Delano* I plant (1991)	Agricultural waste	Bubbling fluidised bed	29	-
CP	*McNeil* Plant (1984)	Wood (unspecified)	Travelling grate	30	-
CP	*Enstedvaerket* (1998)	Straw, wood chips (0-20%)	Shredded straw/stoker; wood chips are burned in a separate boiler to super-heat the steam from straw	41*	-
CHP	*Handelovaerket* (1994)	MSW, industrial waste, waste wood, sludge	Circulating fluidised bed	13	77
CHP	*Masnedoe* (1996)	Straw, wood chips	oscillating grate; Shredded straw/stoker	26	91
CHP	*Vejen* (-)	Waste, straw, wood chips	Sectional step grate for waste and wood chips; cigar burner for straw	21*	83
CHP	*Maabjerg* (1993)	Waste, straw, nat. gas, wood chips	Vibrating grate for waste; cigar burner for straw and wood chips	27	92
CHP	*Oestkraft* (1995)	Wood chips (20%), coal (80%), oil	Travelling grate; Woodchips are substituted with oil when the boiler loads > 65% of the boiler nominal	35	88
CHP	*Hjordkær* (1997)	Wood chips, biowaste	Step grate; pre-combustor; initially used as pilot plant	16*	86^{α}
CHP	*Assens* (1999)	Wood chips, mix (wood waste, residues)	Pneumatic feeders; oscillating grate	25	85^{β}
CHP	*Rudkoebing* (1990)	Straw	Shredded straw/stoker	21	85
CHP	*Haslev* (1989)	Straw	Cigar burner	23	83
CHP	*Slagelse* (1990)	Straw	Shredded straw/stoker	27	92
CHP	*Grenaa* (1992)	Straw	Circulating fluidized bed; Shredded straw/pneumatic feeder	18^{γ}	76
CHP	*Maribo/Sakskoebing* (2000)	Straw	Shredded straw/stoker	29	94
CHP	*Alholmens Kraft* (-)	Waste wood, forest residues, coal, oil, peat	Circulating fluidized bed	37	57
CHP	*Herning* (2009)	Wood chips (70%), wood pellets (30%),	water-cooled vibration grate; pneumatic spreaders	28	87
CHP	*Avedoerevaerket* (Block 2) (2001)	Straw, wood pellets, fossil fuel	Separate straw (ultrasupercritical) boiler; wood pellets are milled and fed together with coal; one common steam turbine	$41^{\dagger}$	$93^{\dagger}$

*Gross efficiency.

α Designed to supply primarily district heating.

β Without flue-gas condensation. The η_{tot} is between 93%-97% including flue-gas condensation.

γ Low electricity production as the plant was designed to supply primarily process steam to industry.

†Full-load efficiency (CHP mode). The annual average electricity (as well as heat) efficiency is 38%. In condensing mode the electricity efficiency can be up to 49%.

Table S7. Air emissions (only main chemicals) from biomass and bio/syngas combustion (44). Values are expressed per GJ of primary energy (LHV_{wb}, i.e., LHV wet basis) of the fuel combusted. $PCDD/F^-$: dioxins and furans (as Polychlorinated Dibenzo-p-dioxins, i.e., PCDDs); TSP: total suspended particulate; UHC: unburned hydrocarbons.

Air emission	Unit	Biogas fuelled engines	Syngas fuelled engines	Straw combustion	Wood combustion
CO	$g\ GJ^{-1}$	310	586	67	90
CH_4	$g\ GJ^{-1}$	434	13	<0.47	<3.1
N_2O	$g\ GJ^{-1}$	1.6	2.7	1.1	0.83
NO_x	$g\ GJ^{-1}$	202	173	125	81
$PCDD/F^-$	$ng\ GJ^{-1}$	<0.96	<1.7	<19	<14
HCl	$g\ GJ^{-1}$	-	-	56	-
Naphthalene	$\mu g\ GJ^{-1}$	4577	8492	12088	2314
NMVOC	$g\ GJ^{-1}$	10	2.3	<0.78	<5.1
∑PAH	$\mu g\ GJ^{-1}$	<606	<181	<5946	<664
SO_2	$g\ GJ^{-1}$	-	-	49	<1.9
TSP	$g\ GJ^{-1}$	-	-	<2.3	10
UHC	$g\ GJ^{-1}$	333	12	<0.94	<6.1

3.5 Transportation

Transportation of the harvested biomass from the farm to the energy plant was included in the model. A transportation distance of 50 km was assumed. Since the three crops were assumed to have similar water content after drying and storage, the fuel consumption for transport was based on the data provided by (32) for on field dried straw bales (similar water content). Transportation of the digestate from the anaerobic digestion plant to the field of application was not included in the modeling.

3.6 Treatment of thermal conversion residues

Bottom ash from gasification, combustion and co-firing scenarios was assumed to be used for road construction substituting for extraction and production of gravel, following the approach of (73). Recovery of phosphorous from the bottom ashes was not included; although this might be an option in the future, at this stage of the research the authors are not aware of established and available technologies for P extraction from the bottom ashes. The fly ashes were assumed to be disposed of in an old salt mine with negligible environmental impacts. Treatment of waste water was not included either in the LCA model.

3.7 Digestate storage

The emission of CH_4 from digestate storage was calculated using the same approach as for crop storage (section 3.2). The emission of biogenic CO_2 was estimated as for the biogas, i.e., as a function of the biogenic CH_4 releases, based on the methodology described by (8). The N losses during the storage of the digestate were estimated using the same approach as for the N losses from crop storage. The losses flows are illustrated in Figure S13-S18.

3.8 Use on land of digestate

The amount and composition of the digestate derived from anaerobic digestion of the crops was calculated based on a mass balance approach, i.e., as the difference between the initial nutrients and dry matter fed to the digestion process and the amount transferred to the biogas, considering the subsequent losses occurring during the digestate storage. The digestate from anaerobic digestion was assumed to substitute for N, P and K mineral fertilizers, considering the digestate is fertilizing the 6 years rotation of winter barley, winter rape, winter wheat (twice) and spring barley (twice) described in (8), for a pig farm. Fertilizer substitution is further detailed in section 6 of this SI.

The emission and leaching of nutrients were quantified as follows: direct N_2O emissions were calculated equal to 1.5% of the N applied with the digestate based the mean value of the range provided by the IPCC approach (74) for application on land of digestate. The emission of NH_3-N was calculated equal to 11% which is the average of a range of values (Table S17) suggested by (75-78) (results in Table S8). The influence of the uncertainty associated with these values on the LCA results was assessed in the uncertainty analysis. The leaching of N (as nitrates) was calculated equal to 45% of the digestate N content based on (8). The indirect N_2O emission (i.e., N_2O produced from secondary reactions involving NH_3, NO_x and leached N) were quantified based on IPCC (74). With respect to this, the N_2O flows associated with use on land shown in Figure S16-S18 only refer to the direct N_2O emissions. Losses of P to soil and water were considered to correspond to 5% of the P applied in excess, based on (9). The K losses to soil and water were not further considered, as not affecting the environmental categories considered, based on the impact assessment methodology selected.

The share of the applied C that enters the soil C pool and that is emitted as CO_2 was determined based on the findings of (78). Based on this, it was considered that 66% of the initial C applied is emitted as CO_2 after 1 year, and 74% after 20 year (Figure S13-S15 and Table S8).

4. Carbon and nitrogen flows

As described in the main manuscript (section 2.4) the C and N flows of all the scenarios assessed in this study have been disaggregated and calculated for all the major processes involved. This included the soil C changes resulting from the cultivation stage, which were calculated with the dynamic soil C model C-TOOL (79, 80), as detailed in (9) for all crop systems. The modeling of the other C and N flows was based on the equations listed in section 9 of this document. The carbon and nitrogen flow analysis was facilitated with the software STAN (81). The values reported in the sankey-flows refer to calculated mean value (eventually reconciliated by STAN) with relative standard deviation. The C and N flows for ryegrass, willow and *Miscanthus* are reported in Figure S13-S18.

Table S8 summarizes the major C and N flows for all the 12 bioenergy scenarios assessed. The discussion of the results can be found in the main manuscript (section 3.1).

Table S8. Overview of (selected) carbon (t C ha^{-1} y^{-1}) and nitrogen (kg N ha^{-1} y^{-1}) flows in the bioenergy scenarios (rounded average values); C atm: carbon uptake from atmosphere; CO_2-C: carbon released during field (including C uptaken from atmosphere and agronomic inputs, and not embedded in the harvestable products and residues) and energy processes; ΔSOC: change in soil organic carbon; CO_2-$C_{avoided}$: avoided carbon emission due to (fossil) energy substitution. Negative values represent inflows, sinks and avoided emissions (e.g., uptake, ΔSOC, etc.).

		Ryegrass				Willow				*Miscanthus*			
Phase	Emission	AD	GA	CO	CF	AD	GA	CO	CF	AD	GA	CO	CF
Cultivation	C atm	-12				-11				-11			
	CO_2-C	6.9				6.1				6.4			
	ΔSOC	-0.51				-0.53				-0.48			
	N leached	74				10				10			
	N_2-N	58				23				20			
	NH_3-N	47				24				6.6			
	N_2O-N	5.8				2.3				2.0			
	NO_x-N	6.8				2.3				1.7			
Energy use	CO_2-C	3.2	4.5	4.7	4.7	3.6	5.6	6.0	6.0	2.9	4.2	4.5	4.5
	CH_4-C	0.049	0.002	-	-	0.053	0.002	-	-	0.041	0.002	-	-
	CO_2-$C_{avoided}$	-4.6	-4.6	-5.7	-6.6	-4.9	-5.9	-7.1	-8.3	-3.9	-4.5	-5.9	-6.9
Digestate use on land	CO_2-C	2.4	-	-	-	3.5	-	-	-	2.5	-	-	-
	C in soil	-1.2	-	-	-	-1.7	-	-	-	-1.3	-	-	-
	NH_3-N	71	-	-	-	58	-	-	-	43	-	-	-
	N_2O-N(dir.)	9.7	-	-	-	7.9	-	-	-	5.9	-	-	-
	N_2O-	3.3	-	-	-	2.4	-	-	-	1.8	-	-	-
	NO_3^--N	290	-	-	-	240	-	-	-	180	-	-	-
	N in soil	270	-	-	-	220	-	-	-	160	-	-	-

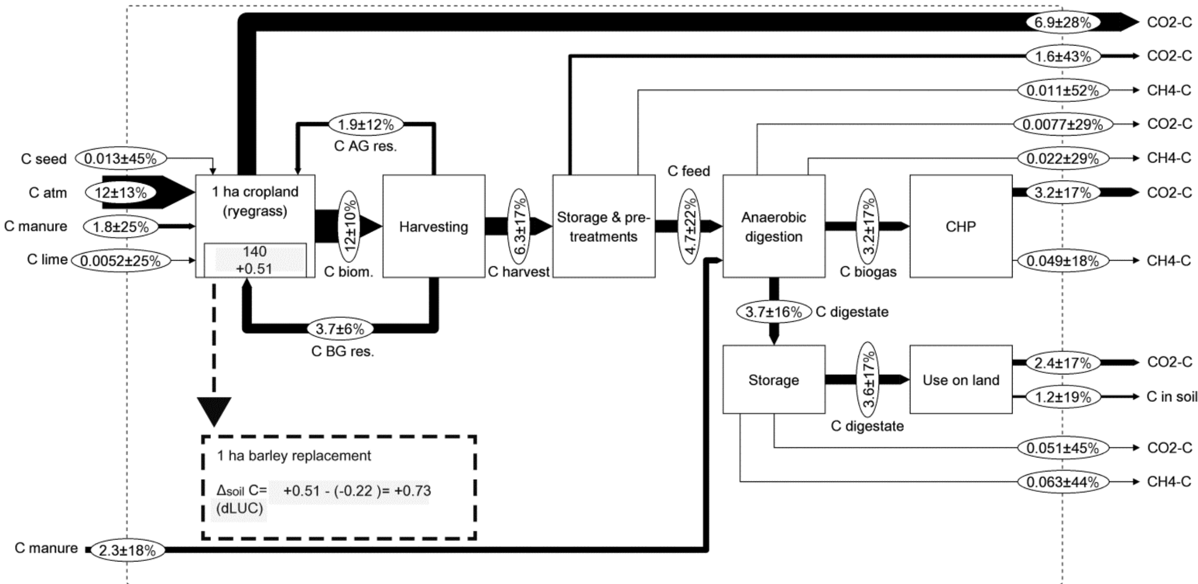

Figure S13. Illustration of the C flows breakdown (t C ha^{-1} y^{-1}) for anaerobic co-digestion of ryegrass with raw pig manure (values rounded to 2 significant digits). AG stands for above-ground residues and BG stands for below-ground residues. Carbon fossil emissions associated with machinery used in the cultivation, transport, storage and energy use phase are not reported to simplify the diagram; however, these were accounted for in the LCA model.

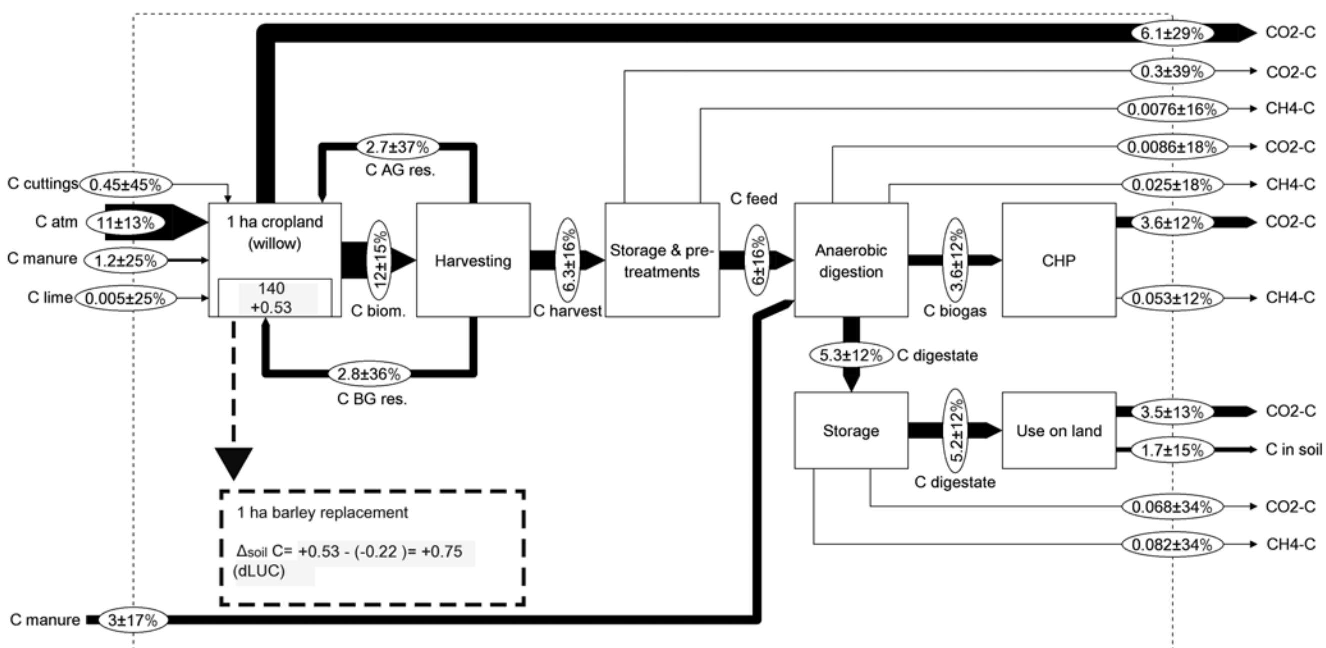

Figure S14. Illustration of the C flows breakdown (t C ha^{-1} y^{-1}) for anaerobic co-digestion of willow with raw pig manure (values rounded to 2 significant digits).

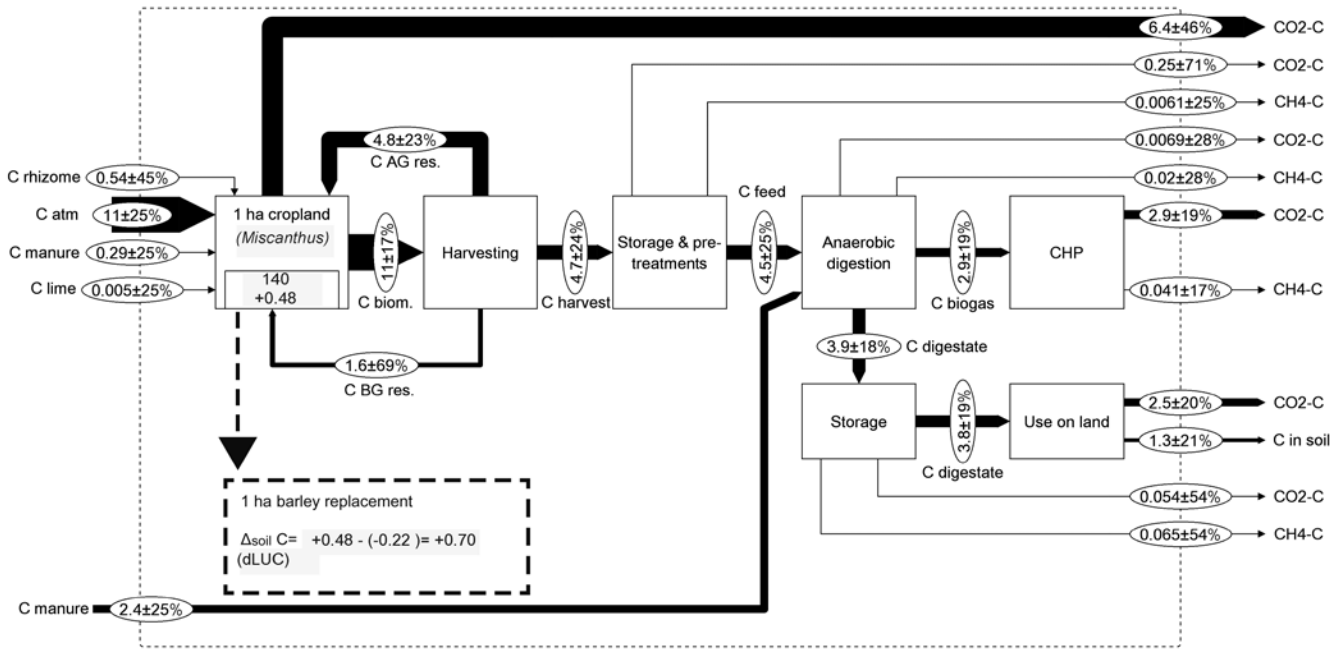

Figure S15. Illustration of the C flows breakdown (t C ha^{-1} y^{-1}) for anaerobic co-digestion of *Miscanthus* with raw pig manure (values rounded to 2 significant digits).

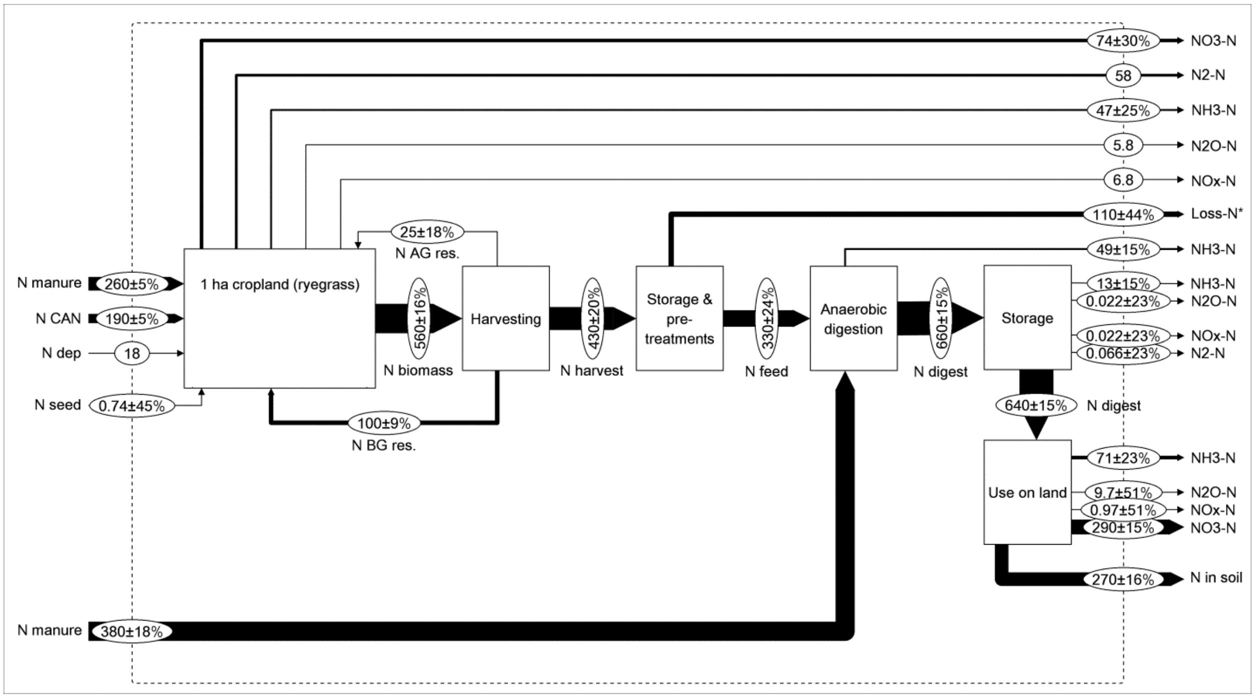

Figure S16. Illustration of the N flows breakdown (kg N ha^{-1} y^{-1}) for anaerobic co-digestion of ryegrass with raw pig manure (values are rounded to 2 significant digits). AG and BG stands for above- and below- ground residues; N* stands for total unspecified N losses during crop storage; indirect N_2O emissions are not illustrated; N in soil also includes eventual N_2 losses.

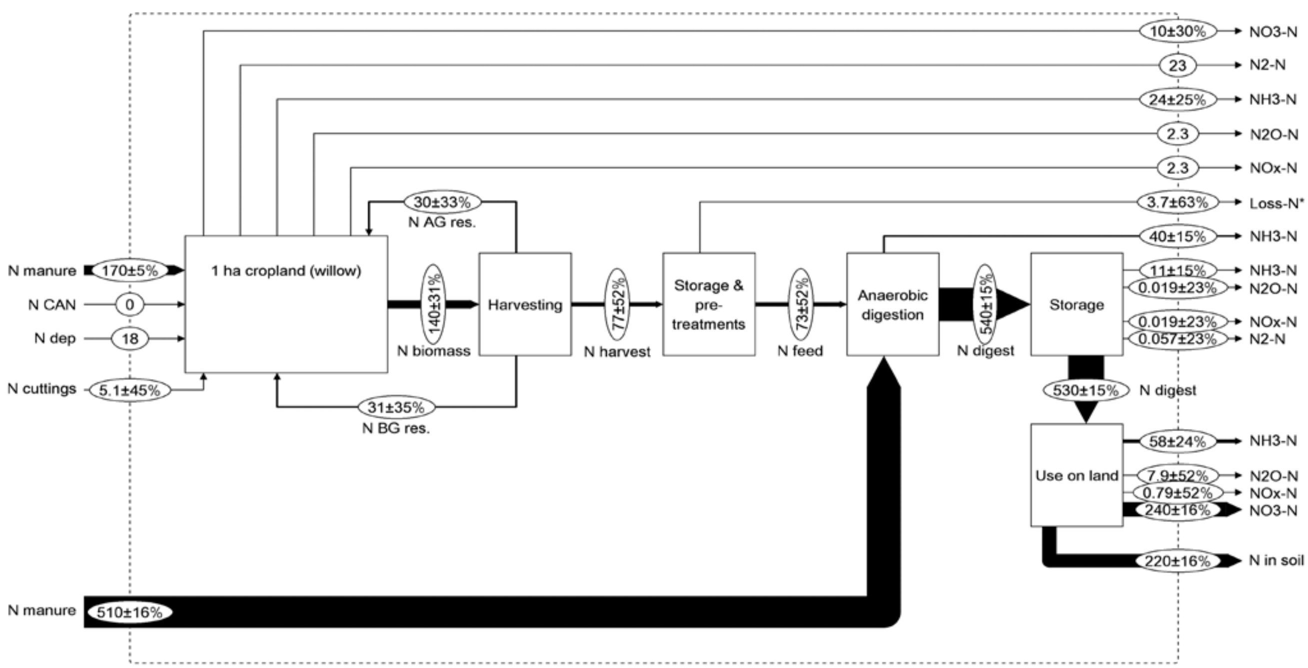

Figure S17. Illustration of the N flows breakdown (kg N ha^{-1} y^{-1}) for anaerobic co-digestion of willow with raw pig manure (values rounded to 2 significant digits).

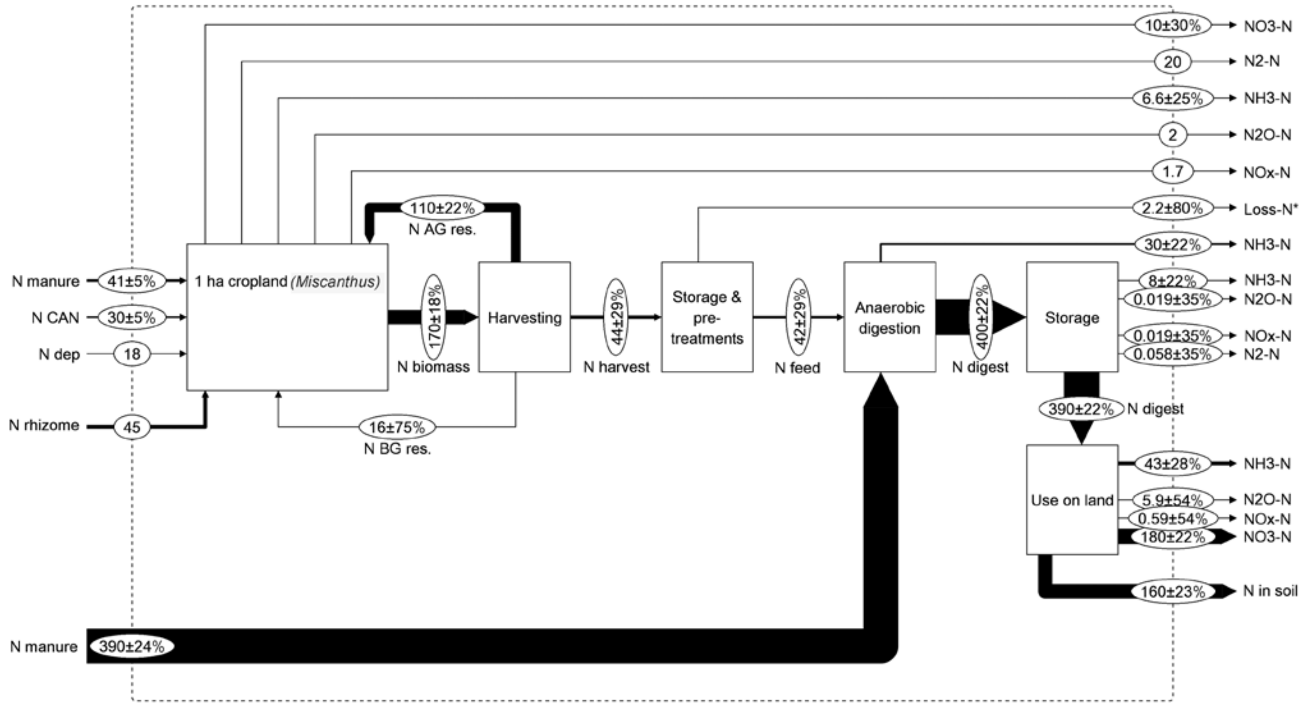

Figure S18. Illustration of the N flows breakdown (kg N ha^{-1} y^{-1}) for anaerobic co-digestion of *Miscanthus* with raw pig manure (values rounded to 2 significant digits).

5. Energy balance of the bioenergy scenarios

The energy balance of the 12 bioenergy scenarios assessed is presented in Table S9.

Table S9. Overview of the energy balance of the 12 bioenergy scenarios (rounded average values); db: dry basis (i.e., the value is based on the LHV_{db}); wb: wet basis (i.e., the value is based on the LHV_{wb}); η_{el}: electricity efficiency; η_{ht}: heat efficiency; η_{tot}: total efficiency crop to energy, calculated by dividing the final net electricity and heat produced by the initial energy yield (dry basis). For combustion (CO) and co-firing (CF) the efficiency reported (η_{el}) is a net efficiency (i.e., plant own consumption is subtracted).

		Ryegrass				Willow				*Miscanthus*			
		AD	GA	CO	CF	AD	GA	CO	CF	AD	GA	CO	CF
Cultivation	Yield (t DM ha^{-1} y^{-1})	14 (13.6)				13 (12.7)				10			
	Yield (t FM ha^{-1} y^{-1})	77				25				11			
	$Energy_{db}$ (GJ ha^{-1} y^{-1})	230				230				180			
	$Energy_{wb}$ (GJ ha^{-1} y^{-1})	77				200				180			
Pretreatment	El. (MWh ha^{-1} y^{-1})	0.1	0.1	-	0.1	0.1	0.1	-	0.1	0.1	0.1	-	0.1
	Heat (GJ ha^{-1} y^{-1})	-	-	-	-	16	-	-	-	12	-	-	-
	DM loss (t DM ha^{-1} y^{-1})	3.3				0.6				0.6			
Operation	El. (MWh ha^{-1} y^{-1})	0.78	0.3	$4.6^{\dagger}$	$4.6^{\dagger}$	0.9	0.3	$5.1^{\dagger}$	$5.1^{\dagger}$	0.7	0.7	$6.1^{\dagger}$	$6.1^{\dagger}$
	Heat (GJ ha^{-1} y^{-1})	9.3	-	-	-	12	-	-	-	9.4	-	-	-
Crop fed	Crop fed (t DM ha^{-1} y^{-1})	10				12				9.4			
	Crop fed (t FM ha^{-1} y^{-1})	12				14				11			
	$Energy_{db}$ (GJ ha^{-1} y^{-1})	170				220				170			
	$Energy_{wb}$ (GJ ha^{-1} y^{-1})	170				210				170			
Raw pig manure	Amount (t DM ha^{-1} y^{-1})	4.7	-	-	-	6.3	-	-	-	5.0	-	-	-
	Amount (t FM ha^{-1} y^{-1})	69	-	-	-	92	-	-	-	72	-	-	-
Gas conversion	$Energy_{gas}$ (GJ ha^{-1} y^{-1})	140	120	-	-	160	150	-	-	130	120	-	-
Energy efficiency	η_{el} (%)	38	38	27	38	38	38	27	38	38	38	27	38
	η_{ht} (%)	52	52	63	52	52	52	63	52	52	52	63	52
Net energy output	El. (MWh ha^{-1} y^{-1})	14	13	13	18	16	16	16	23	13	12	13	19
	Heat (GJ ha^{-1} y^{-1})	65	64	110	88	56	80	140	110	45	62	110	92
η_{tot} (crop-energy)	$\eta_{tot\ el}$ (%)	22	20	20	28	26	25	25	36	26	25	27	38
	$\eta_{tot\ ht}$ (%)	28	28	47	38	24	35	59	48	25	35	62	52

†The electricity consumption is reported although this is already accounted for in the net efficiency reported in the line 'Energy efficiency'.

6. Mineral fertilizer substitution for digestate use on land

As described in the main manuscript, it was considered that the digestate was applied to the 6-year crop rotation described in (8), for a representative Danish pig farm. The P and K requirements of this crop rotation are presented in Table S10. The amount of P and K in all produced digestates is shown in Table S11 (N content is also reported). This was calculated based on the P and K content of each energy crop (Table S1), and on the dry matter (DM) of the digestate which was applied on land (Table S11).

The calculation of the amount of mineral fertilizers substituted from using the digestates as organic fertilizers was based on the Danish law (82). Based on this, the amount of N that can be brought into the field is limited, so the N cannot be applied in excess. However, not all the N applied translates into mineral fertilizer avoided, as the law considers an efficiency of 75% for pig slurry (i.e., 100 kg N from organic fertilizer substitutes 75 kg of mineral fertilizer).

On the other hand, the P and K may be applied in excess, as they are not limited as in the case of N. In cases where these are applied in excess, the amount of mineral P and K fertilizers that are avoided should not include the amount of P and K contributing to the excess (8), the rationale being that without the digestate, farmers would only apply minerals P and K up to the crop requirements, in order to save on costs. The proportion of P and K from the applied digestate that are really avoided is therefore calculated as the ratio between the average annual needs in P and K from the crop rotation considered (Table S10), and the content in P and K in the digestate applied (Table S11). As a result, only 18%, 21% and 18% of the P applied respectively with the digestate derived from co-digestion of manure with ryegrass, willow and *Miscanthus* does correspond to avoided mineral P fertilizers, the rest being an excess that would not have been applied otherwise. Similarly, only 25%, 21% and 23% of the K applied does replace mineral K fertilizers. These figures indicate that for all digestates, the nutrients are applied in excess compared with the average annual crop needs (23 kg P ha^{-1} y^{-1} and 61 kg K ha^{-1} y^{-1}).

The same methodology was applied to calculate the amount of mineral fertilizer that would have been substituted in the case of that the manure was applied on land (reference scenario). Table S12 shows the N, P, K content of the raw pig manure used for co-digestion (instead of directly on land) in the individual bioenergy scenarios, the crops uptake rate and the consequent induced N, P and K fertilizers production.

Table S10. P and K requirements of the 6-year crop rotation on which the digestate is applied.

Year	Crop	P (kg ha^{-1})*	K (kg ha^{-1})*
1	Winter barley	21	54
2	Winter rape	30	89
3	Winter wheat	22	66
4	Winter wheat	22	66
5	Spring barley	22	45
6	Spring barley	22	45
Annual average		23	61

* Data for P and K requirements are from (83).

Table S11. Amount of N, P, K applied and avoided with/from the digestates produced in the individual anaerobic digestion scenarios (values rounded to 2 significant digits).

Bioenergy scenario	Digestate' nutrients (kg ha^{-1})				Uptake (%)			Avoided fertilizers production (kg ha^{-1})		
	DM	N	P	K	N	P	K	N	P	K
AD RG	8900	640	130	240	75	18	25	480	23	61
AD WI	12000	530	110	300	75	21	21	400	23	61
AD MI	7300	390	130	280	75	18	23	290	23	61

Table S12. Amount of N, P, K in the total raw manure used for each individual anaerobic co-digestion scenario and amount of mineral N, P, K fertilizers induced from not applying the raw manure directly on land anymore (values rounded to 2 significant digits). N, P, K content is according to reference values suggested by the Danish legislation for ex-storage raw pig manure (8).

Bioenergy scenario	Nutrients in the total raw manure ab-housing used for AD (kg ha^{-1})				Uptake (%)			Induced fertilizers production (kg ha^{-1})		
	DM	N	P	K	N	P	K	N	P	K
AD RG	4700	330	72	180	75	32	34	280	25	66
AD WI	6300	440	96	240	75	24	26	370	25	66
AD MI	5000	350	75	190	75	32	33	290	25	66

7. Indirect land use changes

In order to evaluate the amount of land expanded per hectare of spring barley displaced from Denmark, the results of (84) have been used, as shown in Table S13. The result of Table S13 corresponded to a total of 0.17 ha expanded per tonne of wheat extra demand (1 ha=10,000 m^2).

In this study, these results were used as a rough approximation for the land expansion due to displacing 1 hectare of spring barley. For this, a yield of 4.9 t DM ha^{-1} was considered for spring barley, based on (9) as well as a DM content of 85%. As a result, 0.95 ha are expanded per ha displaced (0.17 ha expanded t^{-1} wheat (taken as a proxy for barley) × 4.9 t DM t^{-1} barley (fresh) / 0.85 t DM t^{-1} fresh barley). Table S13 shows how the 0.17 ha expanded (per tonne of wheat extra demand) calculated by (84) is distributed among the different regions of the world. The same author also presented these results over an aggregation of 8 regions only, as shown in Table S14. In (84), the results of Table S14 are further translated into affected biomes. This is presented in Table S15. In order to relate the results of Table S14 and S15, Table S16 has been used (taken directly from (84)). Based on the results of Tables S13-S16, Table 1 of the main manuscript could be drawn (i.e., the results from its first fourth columns).

Table S13. Results for 1 t of wheat demand increase from Denmark (values as reported in (84)).

Net expansion (m^2 t^{-1} wheat extra demand)		Australia	Rest of Oceania	China	Rest of E and SE Asia	Japan	Rest of S Asia	India	Middle E and N Africa	Canada	USA	Mexico	Central and rest of N America	Rest of S America	Peru	Brazil	Rest of EU15	EU12	Denmark	Rest of Europe	Former Soviet Union	S African Customs Union	Rest of Sub-Saharan Africa	TOTAL
		aus	xoc	chn	xea	jpn	xsa	ind	xme	can	usa	mex	xca	xla	per	bra	xeu15	eu12	dnk	xer	xsu	xsc	xss	
DK-core	Cult. Land	107.1	11.3	0.0	7.5	1.5	0.0	4.6	33.9	96.9	0.0	15.7	6.1	70.6	9.4	176.2	227.7	0.0	0.0	10.1	91.1	0.0	285.1	
	Graz. Land	37.0	3.3	0.0	6.1	0.0	1.8	1.1	0.0	10.0	67.9	0.0	2.9	16.5	0.3	41.2	133.4	13.7	-8.2	3.9	90.6	0.0	81.4	
	Total	144.1	14.5	0.0	13.6	1.5	1.8	5.7	33.9	106.9	67.9	15.7	9.0	87.0	9.7	217.5	361.1	13.7	-8.2	14.1	181.7	0.0	366.5	
Total - verification		144.1	14.5	0.0	13.6	1.5	1.8	5.7	33.9	106.9	67.9	15.7	9.0	87.0	9.7	217.5	361.1	13.7	-8.2	14.1	181.7	0.0	366.5	1658

Table S14. Results for Denmark aggregated over 8 regions only (from (84)).

Net expansion (m^2 t^{-1} wheat extra demand)	TOTAL	Sub-saharan Africa excluding SACU*	EU-15, excluding Denmark	Brazil	Former Soviet Union, excluding the Baltic States	Australia	Canada	South America, excluding Brazil and Peru	United States of America	Rest of the world
		xss	xeu15	bra	xsu	aus	can	xla	usa	row
Cultivable land	1,155	285	228	176	91	107	97	71	0	100
Grazable land	503	81	133	41	91	37	10	16	68	25
Total	1658	367	361	217	182	144	107	87	68	125

* SACU: South African Customs Union: Botswana, Lesotho, Namibia, South Africa, Swaziland.

Table S15. Results for Denmark, translated into affected biomes (after (84), values rounded to two significant digits).

Biomes	Area converted (per 1 t wheat extra demand)
Savanna	300 m^2
Tropical evergreen forest	350 m^2
Boreal deciduous forest	97 m^2
Evergreen/deciduous mixed forest	200 m^2
Dense shrubland	260 m^2
Grassland/steppe	150 m^2
Open shrubland	170 m^2
Boreal evergreen forest	10 m^2
Rest (biomes unknown)	130 m^2
Total	1700 m^2

Table S16. Correspondence between the region and biomes affected (from (84)).

Region	Biomes affected on cultivable land	Biomes affected on grazable land
Aus	Savanna	Open shrubland & grassland/steppe
Bra	Tropical evergreen forest	savanna
Can	Boreal deciduous forest	Boreal evergreen forest
Xeu15	Evergreen/deciduous mixed forest & dense shrubland	Dense shrubland
Xsu	Grassland/steppe	Evergreen/deciduous mixed forest
Xla	Grassland/steppe & tropical evergreen forest	Savanna & dense shrubland
Xss	Tropical evergreen forest & savanna	Open shrubland
usa	(full utilization of cultivable land)	Open shrubland

8. Sensitivity and uncertainty analysis

As illustrated by (85), uncertainties in LCA studies can generally be distinguished as: I) model uncertainties, II) scenario uncertainties and III) parameter uncertainties. The first is associated with the models and equations used to quantify the different substance flows and with the impact assessment methodology selected which provides the characterization factors for relating the inventoried substances to environmental impacts. Scenario uncertainties is related to uncertainties associated with the choice of technologies and processes and to the fundamental assumptions intrinsically connected to the consequential LCA approach, that is, the choice of the marginal crop and energy production technologies replaced in the market by the modeled cascading effects. Finally, parameter uncertainties reflect the uncertainty intrinsically associated with life cycle inventory data (e.g., in this study: crop yield, crop properties, energy efficiency of the BtE technologies, etc.).

The approach used in this study was as follows: I) model and equation uncertainties were not addressed as these were basic mathematical equations and mass/energy balances (see section 9). The uncertainty of the characterization factors was also not assessed as this was out of the scope of the paper and as the uncertainty of the methodology equally applies to all the selected bioenergy scenarios. II) Scenario uncertainty was tested for the most influencing assumptions; a) variation (min-max) of the iLUC impacts with respect to CO_2 emissions (vs. mean value assumed for the baseline); b) winter wheat as the marginal crop for Denmark (vs. spring barley as for the baseline); c) coal-based heat production as the marginal energy technology for heat generation (vs. natural gas-based as for the baseline); d) natural gas power plant as the marginal technology for electricity generation (vs. condensing coal power plant as for the baseline); e) mono-digestion of the crops (vs. baseline which was based on co-digestion with manure). This scenario illustrates the environmental performance of mono-digestion, that is, excluding the savings associated with raw manure management; f) pre-treatment of pelletization before co-firing (vs. 'no pelletization' as for the baseline). Each of these changes was individually tested to assess the influence of each single change on the overall LCA results. The results of the sensitivity analyses ('a' to 'f') are presented in Figure S19. III) The influence of the parameters uncertainty on the LCA results was tested with a MonteCarlo analysis (number of simulations: 1000; normal distribution assumed). This was done by collecting a set of uncertainties for the most relevant parameters adopted in the model (Table S17). These were the parameters which variation affected the overall energy production of a given bioenergy scenarios (e.g., crop yield, crop properties, energy efficiency, etc.). The approach used to define the uncertainty was as follows: I) the mean and standard deviation was provided for the parameter of interest by the referenced source: in this case the standard deviation was used as such in the model; this was the case for the crops properties (e.g., DM, C, N, K, P and LHV). II) The standard deviation was not directly provided by the referenced source but could, however, be recalculated based on the published values: in this case the standard deviation was quantified based on the available set of values. III) A mean value was reported, whereas the standard deviation for the parameter of interest was not provided; however, a range (max-min) was reported: in this case a normal distribution around the mean value was assumed and the range max-min was assumed equal to the 95% confidence interval; the standard deviation was consequently estimated (i.e., range divided by 4). Table S17 provides an overview of the type of approach (I, II or III) used for the calculation of the mean and relative standard deviation for the parameters selected for the MonteCarlo analysis. The MonteCarlo analysis compared the individual bioenergy scenarios across each other (e.g., ‘A’: combustion of willow vs. ‘B’: combustion of *Miscanthus*). The result of the analysis provided the number of occurrences where the bioenergy scenario ‘A’ allowed for more environmental benefits than ‘B’ on the selected impact category. The results are presented in Table S18 with respect to the environmental category global warming (the analysis was performed only for the relevant combinations of bioenergy scenarios).

Table S17. Overview of normal probability distributions of the selected parameters (rounded values) used in the MonteCarlo analysis to compare the 12 bioenergy scenarios across each other. In brackets the uncertainty range corresponding to the 95% confidence interval (i.e., the interval of length equal to four times the standard deviation around the mean) is reported. CO_2-C atm: carbon uptake from atmosphere; CO_2-C: carbon released during field processes (i.e., not entering the soil C pool); drying loss: dry matter losses from drying on field; CGE: cold gas efficiency; CCE: carbon conversion efficiency; η_{el}: electricity efficiency; η_{ht}: heat efficiency; GE: gas engine; CO: combustion; CF: co-firing; CO_2-C dig: C released after digestate application on land; NH_3-N, N_2O-N: N-emissions in use on land; $Energy_{wb}$: energy of the crop (wet basis) as fed into the energy plant.

Parameter	Unit	RG	WI	MI	Approach	Reference[†]
Yield	t DM ha^{-1}	13.6 (±4.5)	12.7 (±4)	10 (±3.3)	II (RG)/III	3.1
C atm	t CO_2-C ha^{-1}	12 (±3)	11 (±2.9)	11 (±2.9)	I	4
CO_2-C	t CO_2-C ha^{-1}	6.9 (±7)	6.1 (±3.6)	6.4 (±3.7)	I	4
C content	% DM	46.4 (±2.2)	48.9 (±1)	47.7 (±1)	I	3.1
N content	% DM	2.9 (±0.3)	0.6 (±0.3)	0.44 (±0.13)	I	3.1
LHV	MJ kg^{-1} DM	16.8 (±2.4)	18.1 (±0.8)	17.8 (±0.6)	I	3.1
CH_4 yield	% CH_4 pot	70 (±20)	70 (±20)	70 (±20)	III	3.4.1
Drying loss	% DM	20 (±10)	-	-	III	3.3
Storage loss[α]	% DM	5 (±2.5)	4.8 (±1.3)	5 (±2.5)	III	3.2
CGE	%	70 (±15)	70 (±15)	70 (±15)	III	3.4.2
CCE	%	95 (±4)	95 (±4)	95 (±4)	III	3.4.2
η_{el} (GE)	% $Energy_{gas}$	38 (±4)	38 (±4)	38 (±4)	III	3.4.1-3.4.2
η_{el} (CO)	% $Energy_{wb}$	27 (±2)	27 (±2)	27 (±2)	III	3.4.3
η_{el} (CF)	% $Energy_{wb}$	38 (±3)	38 (±3)	38 (±3)	III	3.4.3
η_{ht} (GE)	% $Energy_{gas}$	52 (±8)	52 (±8)	52 (±8)	III	3.4.1-3.4.2
η_{ht} (CO)	% $Energy_{wb}$	63 (±7)	63 (±7)	63 (±7)	III	3.4.3
η_{ht} (CF)	% $Energy_{wb}$	52 (±8)	52 (±8)	52 (±8)	III	3.4.3
CO_2-C dig	% C applied	74 (±9)	74 (±9)	74 (±9)	III	3.8
NH_3-N	% N applied	11 (±4)	11 (±4)	11 (±4)	III	3.8
N_2O-N	% N applied	1.5 (±1.5)	1.5 (±1.5)	1.5 (±1.5)	III	3.8

† Reference section in the text where the data are presented and discussed.
α Indoor storage of dried biomass.

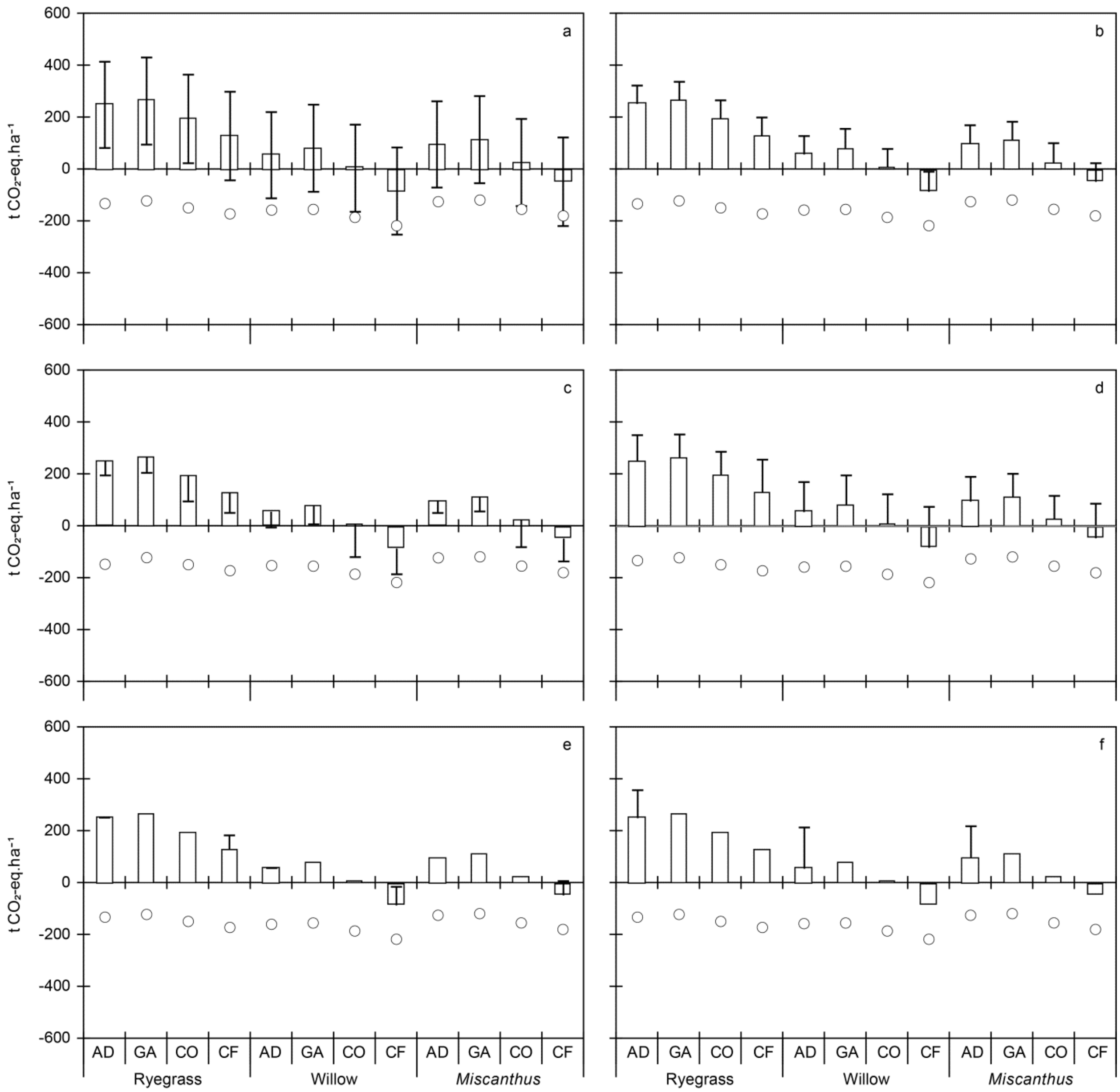

Figure S19. Sensitivity analysis: the error bars illustrate the variation in the LCA results for GW compared with the baseline LCA results. The ‘circle’ indicates the GW saving corresponding to a 35% GHG reduction compared with the reference (used as comparative measure-stick). The following are displayed: a) variation (min-max) of the iLUC impacts with respect to CO_2 emissions (vs. mean value as assumed for the baseline); b) winter wheat as the marginal crop for Denmark (vs. spring barley as for the baseline); c) coal-based heat production as the marginal energy technology for heat generation (vs. natural gas-based as for the baseline); d) natural gas power plant as the marginal technology for electricity generation (vs. condensing coal power plant as for the baseline); e) pre-treatment of pelletization prior to thermal energy conversion (vs. ‘no pelletization’ as for the baseline); f) mono-digestion instead of co-digestion with manure (only applies to the anaerobic digestion scenarios).

Table S18. Uncertainty analysis for global warming based on MonteCarlo analysis: the values indicate the number of occurrences (%) in which the bioenergy scenario 'A' resulted in less environmental impacts than 'B' (e.g., 100 means that 'A' resulted in less impacts than 'B' in 100% of the occurrences).

A<B	B	Ryegrass				Willow				*Miscanthus*			
A		AD	GA	CO	CF	AD	GA	CO	CF	AD	GA	CO	CF
Ryegrass	AD												
	GA	60											
	CO	100	66										
	CF	100	81	59									
Willow	AD	98											
	GA		90			100							
	CO			86		100	77						
	CF				97	100	78	61					
Miscanthus	AD	90				82							
	GA		60				45			100			
	CO			86				43		100	68		
	CF				83				48	100	80	67	

9. List of equations used in the modeling

In this chapter the main equations used for the modeling are listed in order to facilitate the understanding of the carbon and nitrogen flow charts and of the LCA model as well as for sake of transparency.

Emissions during cultivation

Thoroughly detailed in (9).

Emissions from biomass drying

$DL = Yield \cdot K_D$	Eq. S8.
$C\ loss\ drying = DL \cdot C$	Eq. S9.
$N\ loss\ drying = DL \cdot N$	Eq. S10.
$LC_{dr} = C\ loss\ drying/(yield \cdot C)$	Eq. S11.
$LN_{dr} = N\ loss\ drying/(yield \cdot N)$	Eq. S12.

Where:

DL: drying loss	(t DM ha^{-1} y^{-1})
C loss drying: C loss during drying	(t C ha^{-1} y^{-1})
N loss drying: N loss during drying	(t N ha^{-1} y^{-1})
LC_{dr}: C emitted during crop drying as share of initial C	(% C)
LN_{dr}: N emitted during crop drying as share of initial N	(% N)
C: initial carbon content of the crop (at harvest)	(% DM)
N: initial nitrogen content of the crop (at harvest)	(% DM)
Yield: crop yield	(t DM ha^{-1} y^{-1})
K_D: DM loss as share of initial DM	(% DM)

Emissions from biomass storage

$SL = (Yield - DL) \cdot K_L$	Eq. S13.

$$C\,loss\,st = SL \cdot C$$ Eq. S14.

$$N\,loss\,st = SL \cdot N$$ Eq. S15.

$$CH_4 = Yield \cdot CH_{4pot} \cdot 0.67 \cdot MCF$$ Eq. S16.

$$CO_2 = C\,loss\,st - (CH_4 \cdot 12/16) \cdot 44/12$$ Eq. S17.

$$LC_{st} = C\,loss\,st/(yield \cdot C)$$ Eq. S18.

$$LN_{st} = N\,loss\,st/(yield \cdot N)$$ Eq. S19.

Where:

SL: storage loss	(t DM ha^{-1} y^{-1})
C loss st: C loss in storage	(t C ha^{-1} y^{-1})
N loss st: N loss in storage	(t N ha^{-1} y^{-1})
CH_4: emission of methane during storage	(t CH_4 ha^{-1} y^{-1})
CO_2: emission of carbon dioxide during storage	(t CO_2 ha^{-1} y^{-1})
LC_{st}: C emitted during crop storage as share of initial C	(% C)
LN_{st}: N emitted during crop storage as share of initial N	(% N)

DL: drying loss	(t DM ha^{-1} y^{-1})
Yield: crop yield	(t DM ha^{-1} y^{-1})
K_L: loss as share of initial DM	(% DM)
CH_{4pot}: methane potential	(Nm^3 CH_4 t^{-1} DM)
MCF: methane conversion factor	(% CH_{4pot})
K: ratio CO_2/CH_4 in biogas emitted	(without unit)
C: initial carbon content of the crop	(% DM)
N: initial nitrogen content of the crop	(% DM)

The methane conversion factor MCF was estimated equal to 0.5% for biomass storage and 1% for digestate storage. The value 0.5% was based on the MCF suggested for compost storage and the value 1% was based on the MCF for liquid digestate suggested by (30). The coefficient 0.67 is the conversion factor of m^3 CH_4 to kg CH_4 (CH_4 density at 20°C). The ratio 12/16 is the conversion factor between methane and carbon emissions (i.e., kg C kg^{-1} CH_4). The coefficient

K was based on the content of protein, lipid, cellulose, hemicellulose and lignin of the crops and was calculated equal to 2.6 for ryegrass, 2.7 for willow and 2.8 for *Miscanthus*.

Electricity and heat production

Anaerobic digestion:

$$\mathrm{El} = \mathrm{Crop\ fed} \cdot \mathrm{VS} \cdot \mathrm{CH}_{4\,\mathrm{pot}} \cdot \mathrm{CH}_{4\,\mathrm{yield}} \cdot \mathrm{LHV}_{\mathrm{CH_4}} \cdot \eta_{\mathrm{el}}/3.6 \qquad \text{Eq. S20.}$$

$$\mathrm{Ht} = \mathrm{Crop\ fed} \cdot \mathrm{VS} \cdot \mathrm{CH}_{4\,\mathrm{pot}} \cdot \mathrm{CH}_{4\,\mathrm{yield}} \cdot \mathrm{LHV}_{\mathrm{CH_4}} \cdot \eta_{\mathrm{th}} \qquad \text{Eq. S21.}$$

$$\mathrm{Crop\ fed} = \mathrm{yield} - \mathrm{DL} - \mathrm{SL} \qquad \text{Eq. S22.}$$

Gasification:

$$\mathrm{El} = \mathrm{Crop\ fed} \cdot \mathrm{CGE} \cdot \eta_{\mathrm{el}}\ /3.6 \qquad \text{Eq. S23.}$$

$$\mathrm{Ht} = \mathrm{Crop\ fed} \cdot \mathrm{CGE} \cdot \eta_{\mathrm{th}} \qquad \text{Eq. S24.}$$

$$\mathrm{Crop\ fed} = \mathrm{yield} - \mathrm{DL} - \mathrm{SL} \qquad \text{Eq. S25.}$$

Combustion and co-firing:

$$\mathrm{El} = \mathrm{Crop\ fed} \cdot \mathrm{LHV}_{\mathrm{wb}} \cdot \eta_{\mathrm{el}}\ /3.6 \qquad \text{Eq. S26.}$$

$$\mathrm{El} = \mathrm{Crop\ fed} \cdot \mathrm{LHV}_{\mathrm{wb}} \cdot \eta_{\mathrm{th}} \qquad \text{Eq. S27.}$$

$$\mathrm{Crop\ fed} = \mathrm{yield} - \mathrm{DL} - \mathrm{SL} \qquad \text{Eq. S28.}$$

Where:

El: electricity produced	(MWh ha^{-1} y^{-1})
Ht: heat produced	(GJ ha^{-1} y^{-1})
Crop fed: crop fed to the energy plant	(t DM ha^{-1} y^{-1})
Yield: crop yield	(t DM ha^{-1} y^{-1})
SL: storage loss	(t DM ha^{-1} y^{-1})
DL: drying loss	(t DM ha^{-1} y^{-1})
VS: volatile solids	(% DM)
$CH_{4\ pot}$: methane potential	(Nm^3 CH_4 t^{-1} DM)
$CH_{4\ pot}$: methane yield	(% $CH_{4\ pot}$)
LHV_{CH4}: lower heating value of methane (STP)	(35.2 MJ Nm^{-1} CH_4)

LHV_{wb}: Lower heating value of the crop (wet basis) (GJ t^{-1} FM)

η_{el}: net electricity efficiency (%)

η_{th}: net heat efficiency (%)

Carbon dioxide emissions from BtE conversion

Anaerobic digestion:

$$CO_2 = \left[Yield \cdot C \cdot (1 - LC_{dr} - LC_{st}) \cdot CC - E_{C,f} - E_{CCH4}\right] \cdot 44/12 \qquad \text{Eq. S29.}$$

$$CC = \frac{VS \cdot CH_{4\,pot} \cdot CH_{4\,yield}}{(\%CH_4 \cdot V_m)} \cdot \frac{1}{10^3} \cdot \frac{C_m}{C} \qquad \text{Eq. S30.}$$

Gasification:

$$CO_2 = \left[Yield \cdot C \cdot (1 - LC_{dr} - LC_{st}) \cdot CCE - E_{CCH4}\right] \cdot 44/12 \qquad \text{Eq. S31.}$$

Combustion and co-firing:

$$CO_2 = \left[Yield \cdot C \cdot (1 - LC_{dr} - LC_{st}) - E_{CCH4}\right] \cdot 44/12 \qquad \text{Eq. S32.}$$

Where:

CO_2: carbon dioxide produced (t CO_2 ha^{-1} y^{-1})

CC: fraction of carbon biogasified (% C)

LC_{st}: C emitted during biomass storage as share of initial C (% C)

LC_{dr}: C emitted during biomass drying as share of initial C (% C)

Yield: crop yield (t DM ha^{-1} y^{-1})

C: initial carbon content of the crop (% DM)

$CH_{4\,pot}$: methane potential (Nm^3 CH_4 t^{-1} VS)

$CH_{4\,yield}$: methane yield (% $CH_{4\,pot}$)

$\%CH_4$: share of methane in the biogas (65%)

V_m: molar volume of gases (22.414 NL mol^{-1})

C_m: molar weight of carbon (kg mol^{-1})

VS: volatile solids content (% DM)

CCE: carbon conversion efficiency (fraction of C gasified) (% C)

$E_{C,f}$: fugitive emission of biogas (t C ha^{-1} y^{-1})

E_{CCH4}: emission of unburned methane (t C ha^{-1} y^{-1})

The ratio 44/12 is the conversion factor between CO_2 and C emissions (i.e., kg CO_2 kg^{-1} C). The fugitive emission of methane from the digestion plant was set equal to 1% of the methane produced. The emission of unburned methane (E_{CCH4}) can be recalculated from Table S8.

Emissions from digestate storage

$$CH_4 = Yield_{dig} \cdot CH_{4pot} \cdot 0.67 \cdot MCF$$ Eq. S33.

$$CO_2 = CH_4 \cdot K$$ Eq. S34.

$$C\ loss\ dig = (CH_4 \cdot 12/16 + CO_2 \cdot 12/44)$$ Eq. S35.

$$LC_{dig\ st} = C\ loss\ dig/(yield \cdot C)$$ Eq. S36.

$$N\ loss\ dig = C\ loss\ dig/C \cdot N$$ Eq. S37.

$$LN_{dig\ st} = N\ loss\ dig/(yield \cdot N)$$ Eq. S38.

Where:

CH_4: emission of methane (t CH_4 ha^{-1} y^{-1})

CO_2: emission of carbon dioxide (t CO_2 ha^{-1} y^{-1})

C loss dig: C loss in digestate storage (t C ha^{-1} y^{-1})

$LC_{dig\ st}$: C emitted during digestate storage as share of initial C (% C)

N loss dig: N loss in digestate storage (t N ha^{-1} y^{-1})

$LN_{dig\ st}$: N emitted during digestate storage as share of initial N (% N)

$Yield_{dig}$: amount of digestate (t DM ha^{-1} y^{-1})

CH_{4pot}: methane potential (Nm^3 CH_4 t^{-1} DM)

MCF: methane conversion factor (% $CH_{4\ pot}$)

K: ratio CO_2/CH_4 in biogas emitted (without unit)

Yield: crop yield (t DM ha^{-1} y^{-1})

C: initial carbon content of the crop (% DM)

N: initial nitrogen content of the crop (% DM)

A methane conversion factor (MCF) of 1% is used for digestate storage, based on the MCF for liquid digestate suggested by IPCC. With respect to the coefficient 0.67 and K, see earlier explanations.

Emissions from use on land of the digestate from anaerobic digestion

$$CO_2 = \text{Yield} \cdot C \cdot (1 - CC - LC_{dr} - LC_{st} - LC_{st\,dig}) \cdot 0.74 \cdot 44/12 \qquad \text{Eq. S39.}$$

$$NH_3 = \text{Yield} \cdot N \cdot (1 - NC - LN_{dr} - LN_{st} - LN_{st\,dig}) \cdot 0.11 \qquad \text{Eq. S40.}$$

$$NO_3 = \text{Yield} \cdot N \cdot (1 - NC - LN_{dr} - LN_{st} - LN_{st\,dig}) \cdot 0.45 \qquad \text{Eq. S41.}$$

$$N_2O\ \text{direct} = \text{Yield} \cdot N \cdot (1 - NC - LN_{dr} - LN_{st} - LN_{st\,dig}) \cdot 0.015 \qquad \text{Eq. S42.}$$

$$NO_x = N_2O\ \text{direct} \cdot 0.1 \qquad \text{Eq. S43.}$$

$$N_2O\ \text{indirect} = N\ \text{leached} \cdot 0.0075 + (NH_3 + NO_x) \cdot 0.01 \qquad \text{Eq. S44.}$$

Where:

CO_2: carbon dioxide produced	(t CO_2 ha^{-1} in 20y)
NH_3: ammonia emission	(t N ha^{-1} y^{-1})
NO_3: nitrates leaching	(t N ha^{-1} y^{-1})
NO_x: NO_x emission	(t N ha^{-1} y^{-1})
N_2O: direct: nitrous oxide emission (direct)	(t N ha^{-1} y^{-1})
N_2O: indirect: nitrous oxide emission (indirect)	(t N ha^{-1} y^{-1})

LC_{dr}: C emitted during crop drying as share of initial C	(% C)
LC_{st}: C emitted during crop storage as share of initial C	(% C)
$LC_{st\,dig}$: C emitted during digestate storage as share of initial C	(% C)
CC: fraction of carbon biogasified	(% C)
LN_{dr}: N emitted during crop drying as share of initial N	(% N)
LN_{st}: N emitted during crop storage as share of initial N	(% N)
$LN_{st\,dig}$: N emitted during digestate storage as share of initial N	(% N)
NC: nitrogen converted into N in biogas	(% N)

Yield: crop yield (t DM ha^{-1} y^{-1})

C: initial carbon content of the crop (% DM)

N: initial nitrogen content of the crop (% DM)

The coefficient NC was estimated to 7% of the N content based on (31). The coefficient CC was calculated according to Eq. S30. The emission of carbon from digestate application on land (74% of the initial carbon applied after a 20 year period) was recalculated to 66% after 1 year period (see Figure S13-S15).

Other equations

Calculation of the reference EU 35% GHGs emission reduction target

$$\mathrm{GHGs}_{\mathrm{EU\,35\%}} = \mathrm{GHGs}_{\mathrm{fossil\,ref}} \cdot (100\% - 35\%) \qquad \text{Eq. S45.}$$

$$\mathrm{GHGs}_{\mathrm{EU\,35\%\ relative}} = \mathrm{GHGs}_{\mathrm{EU\,35\%}} - \mathrm{GHGs}_{\mathrm{fossil\,ref}} \qquad \text{Eq. S46.}$$

Where:

$\mathrm{GHGs}_{\mathrm{EU\ 35\%}}$: GHGs emission (of the individual bioenergy scenario under assessment) that should be achieved to fulfill the EU directive target (t CO_2-eq. ha^{-1})

$\mathrm{GHGs}_{\mathrm{EU\ 35\%\ relative}}$: GHGs emission (of the individual bioenergy scenario) that should be achieved to fulfill the EU directive target ‘minus’ the GHGs emission of the reference fossil fuel system where the hectare of land is used for spring barley cultivation (t CO_2-eq. ha^{-1})

$\mathrm{GHGs}_{\mathrm{fossil\ ref}}$: GHGs emission of the reference fossil fuel system where the hectare of land is used for spring barley cultivation (t CO_2-eq. ha^{-1}). This corresponds to the GHGs emission associated with the provision of the same amount of electricity and heat produced in the individual bioenergy scenario under assessment

10. GWP time-dependency

Over its 20y time scope, this study involves the release of GHG emissions at different periods. For example, the amount of CO_2 emitted from the cultivation stage (i.e., C from the manure as well as from above- and below-ground residues not entering the soil C pool) varies every year as a new equilibrium is reached in the soil. The N_2O emissions related to fertilization occur every year where there is a fertilization event (years 1 to 19, in the *Miscanthus* case, considering the first year as "year 0", in conformity with (86)). The iLUC occur at the very moment energy crops are cultivated in Denmark (year 0). This is further detailed in Table S19.

As detailed in the main manuscript, the impact of GHG time-dependency was tested for the cultivation of *Miscanthus* (including iLUC), based on the methodology described in (86). Table S19 presents the emissions occurring over the 20y time scope of the study, for two selected processes only: cultivation of *Miscanthus* and iLUC. Based on Table S19, as well as on the GWP factors found in the IPCC methodology for a time horizon of 100y, a total of 54705 kg CO_2 eq. ha^{-1} can be calculated for this 20y time period (Table S20). However, using the methodology as well as the calculator provided by (86), a total of 76433 kg CO_2 eq. ha^{-1} is calculated, for this same 20y period, which is ca. 40% higher than the value calculated with the IPCC methodology (Table S20). The reason for this is that the iLUC release, which occurs at year zero, is the most significant CO_2 emission (310 000 kg CO_2eq. ha^{-1}), and also the only one which has the same GWP value with both methods (since it occurs at year 0). After year 0, according to the time-dependency methodology of (86), the later the GHG emissions occur, the smaller their GWP become. In the present case, emissions occurring from year 1 to year 19 correspond to an overall GHG saving (i.e., a negative value). Using the IPCC methodology, this saving would thus be relatively more important than with the method of (86), which explains why the IPCC methodology yields an overall lower GWP result.

Table S19. Annual GHG emissions for the cultivation of *Miscanthus* and iLUC processes.

Year	Cultivation (*Miscanthus*, spring harvest) (sandy loam soil)									iLUC
	soil C	Yearly delta soil C	C manure[b]	C residues[b]	CO_2 manure & residues[c]	CO_2 uptake[b]	CO_2 lime[b]	N_2O (direct)[b]	N_2O (indirect)[b]	CO_2
	(C-TOOL model)[a]	(A)	(B)	(C)	(D) = (B+C-A)*(44/12)	(E)	(F)	(G)	(H)	(I)
	t C ha^{-1}	kg C ha^{-1}	kg C ha^{-1}	kg C ha^{-1}	kg ha^{-1}	kg ha^{-1}	kg ha^{-1}	kg ha^{-1}	kg ha^{-1}	t ha^{-1}
0	144.71									310
1	146.23	1520	144.75	5967	16838	-26499		2.46	0.96	
2	147.32	1090	289.50	6111	19473	-30327		3.04	0.36	
3	148.16	840	289.50	6399	21446	-37983		3.09	0.24	
4	148.70	540	289.50	6399	22546	-37983		3.09	0.24	
5	149.13	430	289.50	6399	22949	-37983		3.09	0.24	
6	149.52	390	289.50	6399	23096	-37983		3.09	0.24	
7	149.89	370	289.50	6399	23169	-37983		3.09	0.24	
8	150.24	350	289.50	6399	23242	-37983		3.09	0.24	
9	150.60	360	289.50	6399	23206	-37983	367.7	3.09	0.24	
10	150.94	340	289.50	6399	23279	-37983		3.09	0.24	
11	151.29	350	289.50	6399	23242	-37983		3.09	0.24	
12	151.63	340	289.50	6399	23279	-37983		3.09	0.24	
13	151.96	330	289.50	6399	23316	-37983		3.09	0.24	
14	152.30	340	289.50	6399	23279	-37983		3.09	0.24	
15	152.63	330	289.50	6399	23316	-37983		3.09	0.24	
16	152.95	320	289.50	6399	23352	-37983		3.09	0.24	
17	153.27	320	289.50	6399	23352	-37983		3.09	0.24	
18	153.59	320	289.50	6399	23352	-37983		3.09	0.24	
19	153.91	320	289.50	6399	23352	-37983		3.09	0.24	
20-99[d]										

[a] The numbers presented in this column are the output from the C-TOOL model, which is detailed in (81) and (82).

[b] Values from (8).

[c] In this study, this emission was considered as 19796 kg ha^{-1} (y1), 20856 kg ha^{-1} (y2), and 22781 kg ha^{-1} (y3-19), as soil C changes were annualized over a 20y period instead of being calculated precisely for each year as in this Table.

[d] Releases from this point are not included as they fall beyond the time scope of the study (20 years).

Table S20. GWP results for the *Miscanthus* cultivation and iLUC processes over the 20 years time scope of the study, with and without accounting for time-dependency (for a time horizon of 100 years).

Total GWP calculated in this study (IPCC AR4 methodology, for 100 years)	(kg CO_2 eq. ha^{-1})	54 705
Total GWP calculated accounting for time-dependency ((86) methodology, for 100 years)	(kg CO_2 eq. ha^{-1})	76 433[a]
Relative difference		40%

[a] This result was obtained from the Excel-based calculator provided as a supporting information by (86).

As shown in Table S20, the global warming results presented in this study could have been relatively higher (ca. 40% for the *Miscanthus* case) if the time-dependency would have been accounted for. This would likely not have changed the ranking observed between the different scenarios, but perhaps the conclusions (i.e., the net overall results in terms of GHG savings or net emission). This emphasizes the research need towards the development of recognized methodologies for reflecting the different GWP of releases occurring at different time periods over the time scope of bioenergy studies.

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