

Appendix A. Supporting Information for Paper I

Appendix 1. *Miscanthus* and willow life cycles

This appendix details the different activities considered throughout each year of both *Miscanthus* and willow life cycles are further detailed (Table S1).

As shown in Table S1, herbicides are, for *Miscanthus*, applied every year until the first harvest (year 1, 2 and 3). Then, one treatment each 4th year to clean up for certain problematic weed species is applied (year 4, 8, 12 and 16). Fertilizers are applied every year except for the first year of the life cycle.

For willow, herbicides are applied on year 1, 2, 3 and 6 (i.e. right after the first harvest) and then a systemic herbicide (glyphosate) is applied every other rotations (i.e. year 12 and 18). Fertilization is performed every year, starting from the year after planting (year 3). Under Danish conditions, slurry spreading is only possible in harvest years, so it is considered that the field is fertilized with 100 % slurry on harvest years, and with 100 % mineral fertilizers for the other years.

Table S1. Summary of activities considered throughout willow and *Miscanthus* life cycles

Year	<i>Miscanthus</i>	Year	Willow
1	Herbicide application. Soil preparation. No fertilization.	1	Herbicide application. Soil preparation. No fertilization.
2	Fertilizer application. Planting. Herbicide application.	2	Herbicide application. Planting.
3	Fertilizer application. First harvest (60 %). Herbicide application.	3	Fertilizer application (100 % mineral). Herbicide application.
4-20	Year 3 repeated, but full harvest. An herbicide treatment each 4 th year (year 4, 8, 12 and 16).	4	Fertilizer application (100 % mineral).
		5	Fertilizer application (100 % mineral).
		6	First harvest. Fertilizer application (100 % slurry). Herbicide application.
		7	Fertilizer application (100 % mineral).
		8	Fertilizer application (100 % mineral).
		9	Second harvest. Fertilizer application (100 % slurry).
		10	Fertilizer application (100 % mineral).
		11	Fertilizer application (100 % mineral).
		12	Third harvest. Fertilizer application (100 % slurry). Herbicide application.
		13	Fertilizer application (100 % mineral).
		14	Fertilizer application (100 % mineral).
		15	Fourth harvest. Fertilizer application (100 % slurry).
		16	Fertilizer application (100 % mineral).
		17	Fertilizer application (100 % mineral).
		18	Fifth harvest. Fertilizer application (100 % slurry). Herbicide application.
		19	Fertilizer application (100 % mineral).
		20	Fertilizer application (100 % mineral).
		21	Final harvest.

Appendix 2. Modeling details for the 9 main agricultural operations involved: soil preparation, propagation, liming, sowing, plant protection, fertilization, irrigation, harvest and transport field-farm.

1. *Danish soil classification*

Chosen as a representative for sandy soils is soil JB3 of the Danish soil classification (see for example Greve, 2011) and chosen as a representative for sandy loam soils is soil JB6 of the Danish soil classification. This appendix as well as the following appendixes will constantly refer to these appellations.

2. *Soil preparation*

Soil preparation, or tillage, includes ploughing (21 cm), harrowing, seedbed harrowing and rolling, for all crops. When straw (spring barley, winter wheat) and beet tops are incorporated, stubble harrowing is performed, as well as for maize. The data for material consumption (or capital goods) related to the different soil preparation operations were taken from the Ecoinvent life cycle inventory database (Ecoinvent Centre, 2011), but the diesel consumption was adjusted based on the norm values presented by Dalgaard *et al.* (2001). As suggested by Dalgaard *et al.* (2001), the norm values are adjusted based on the soil type; by a factor of 1.0 for soil JB6 and 0.9 for soil JB3. The data from the Ecoinvent database include the machinery production, and all processes are described in Nemecek & Kägi (2007). Table S2 presents, for soil preparation operations, the specific Ecoinvent process used as well as the diesel consumption considered, for both sandy and sandy loam soils. The Ecoinvent database considers a specific weight of diesel of 0.84 kg per liter (Nemecek & Kägi, 2007), so this value is used for conversions in liter per hectare. On sandy soils, the harrowing is performed by spring tine harrow and a light seedbed harrowing is performed, while on sandy loam soils, harrowing is performed by rotary harrow and a heavy seedbed harrowing is considered.

Table S2. Modelling details for soil preparation operations

Soil preparation operation	Ecoinvent process used	Diesel consumption	
		Sandy soil (JB3)	Sandy loam soil(JB6)
Ploughing (21 cm), spring [†]	Tillage, ploughing/CH U	18 l ha ⁻¹	20 l ha ⁻¹
Ploughing (21 cm), autumn [‡]	Tillage, ploughing/CH U	20.7 l ha ⁻¹	23 l ha ⁻¹
Harrowing, by spring tine harrow ^β	Tillage, harrowing, by spring tine harrow/CH U	4.76 l ha ^{-1*}	5.29 l ha ^{-1*}
Harrowing, by rotary harrow ^β	Tillage, harrowing, by rotary harrow/CH U	12.32 l ha ^{-1*}	13.69 l ha ^{-1*}
Seedbed harrowing, light ^δ	Tillage, harrowing, by spring tine harrow/CH U	3.6 l ha ⁻¹	4 l ha ⁻¹
Seedbed harrowing, heavy ^δ	Tillage, harrowing, by rotary harrow/CH U	5.4 l ha ⁻¹	6 l ha ⁻¹
Rolling	Tillage, rolling/CH U	1.8 l ha ⁻¹	2.0 l ha ⁻¹
Stubble harrowing [†]	Mulching, CH/U	6.3 l ha ⁻¹	7.0 l ha ⁻¹

* Diesel consumption taken from the Ecoinvent database (and not from Dalgaard *et al.*, 2001). Values include an adjustment based on the soil type, i.e. a factor of 1.0 for sandy loam soils and 0.9 for sandy soils.

† Only when straw is incorporated.

‡ For all crops on JB3, except for winter wheat, spring ploughing is performed. Else, autumn ploughing is performed.

β On JB3, harrowing is performed by spring tine harrow, and on JB6, by rotary harrow.

δ Light seedbed harrowing on JB3, and heavy on JB6.

3. Propagation

Crops from agricultural production need to be propagated, e.g. through seeds (all annual crops and permanent ryegrass), cuttings (willow) or rhizomes (*Miscanthus*). This stage is not systematically included in life cycle assessments and life cycle inventories, and when it is, it is rarely transparent and detailed.

As a first step, the amount of seed, rhizome or cutting needed per ha of land cultivated for each of the selected crop must be determined. This was done mostly based on the Ecoinvent database as well as on some literature data, as shown in table S3. The amount needed for the catch crop was based on ryegrass.

Table S3. Amount of seed, cutting or rhizome needed per ha of land cultivated, for each crops

Crop	Seed, cutting or rhizome needed*	Reference	Comment
Spring barley	0.014484 kg seed per kg (wet) primary yield (72.42 kg seed ha ⁻¹ on JB3 and 82.64 kg seed ha ⁻¹ on JB6 ; see Note 1) [†]	Nemecek & Kägi (2007)	Process “barley grains IP, at farm/CH U”
Winter wheat	0.02591 kg seed per kg (wet) primary yield (171.00 kg seed ha ⁻¹ on JB3 and 207.28 kg seed ha ⁻¹ on JB6; see Note 2) [†]	Nemecek & Kägi (2007)	Process “wheat grains IP, at farm/CH U”
<i>Miscanthus</i>	20 000 rhizomes ha ⁻¹ .	Styles <i>et al.</i> (2008); Atkinson (2009); Styles and Jones (2007)	Planted the second year
Willow	15 000 cuttings ha ⁻¹ .	Personal communication with Jens B. Kjeldsen, Aarhus University (2010). To be seen as a maximal value.	Planted the second year
Sugar beet	2.9405 x 10 ⁻⁵ kg seed per kg (wet) primary yield (1.66 kg seed ha ⁻¹ on JB3 and JB6; see Note 3) [†]	Nemecek & Kägi (2007)	Process “sugar beets IP, at farm/CH U”.
Silage maize	0.00043933 kg per kg (wet) primary yield (17.08 kg seed ha ⁻¹ on JB3 and 16.91 kg seed ha ⁻¹ on JB6; see Note 4) [†]	Nemecek & Kägi (2007)	Process “silage maize IP, at farm/CH U”.
Ryegrass	0.00037449 kg seed per kg (wet) primary yield (20.76 kg seed ha ⁻¹ on JB3 and 18.35 kg seed ha ⁻¹ on JB6. For catch crop, 0.8322 kg seed ha ⁻¹ for both soil types; see Note 5) [†]	Nemecek & Kägi (2007)	Process “Grass from IP, at farm/CH U”
Catch crop	As for ryegrass (0.8322 kg seed ha ⁻¹ on JB3 and JB6; see Note 6) [†]	Nemecek & Kägi (2007)	Process “Grass from IP, at farm/CH U”

* Values shown are the original values as taken from the reference indicated. Values in parenthesis are adapted for the units considered in this inventory.

[†] All data for dry matter (DM) content used in note 1 to note 6 are from Møller *et al.* (2000) and yields (kg DM ha⁻¹) are from Table 2.

Note 1: Spring barley on:

JB3: 0.014484 kg seed/kg (wet) primary yield * kg primary yield/0.85 kg DM * 4.25 * 10³ kg DM/ha = 72.42 kg seed/ha;
 JB6: 0.014484 kg seed/kg (wet) primary yield * kg primary yield/0.85 kg DM * 4.85 * 10³ kg DM/ha = 82.64 kg seed/ha;

Note 2: Winter wheat on:

JB3: 0.02591 kg seed/ kg (wet) primary yield * kg primary yield/0.85 kg DM * 5.61 * 10³ kg DM/ha = 171.00 kg seed/ha;
 JB6: 0.02591 kg seed/ kg (wet) primary yield * kg primary yield/0.85 kg DM * 6.80 * 10³ kg DM/ha = 207.28 kg seed/ha;

Note 3: Sugar beet on:

JB3 and JB6: 2.9405 x 10⁻⁵ kg seed/kg (wet) primary yield * kg primary yield/0.22 kg DM * 12.45 * 10³ kg DM/ha = 1.66 kg seed/ha;

Note 4: Silage maize on:

JB3: $0.00043933 \text{ kg seed/ kg (wet) primary yield} * \text{kg primary yield}/0.31 \text{ kg DM} * 12.05 * 10^3 \text{ kg DM/ha} = 17.08 \text{ kg seed/ha}$;

JB6: $0.00043933 \text{ kg seed/ kg (wet) primary yield} * \text{kg primary yield}/0.31 \text{ kg DM} * 11.93 * 10^3 \text{ kg DM/ha} = 16.91 \text{ kg seed/ha}$;

Note 5: Ryegrass on:

JB3: $0.00037449 \text{ kg seed/ kg (wet) primary yield} * \text{kg primary yield}/0.18 \text{ kg DM} * 9.98 * 10^3 \text{ kg DM/ha} = 20.76 \text{ kg seed/ha}$;

JB6: $0.00037449 \text{ kg seed/ kg (wet) primary yield} * \text{kg primary yield}/0.18 \text{ kg DM} * 8.82 * 10^3 \text{ kg DM/ha} = 18.35 \text{ kg seed/ha}$;

Note 6: Catch crop:

JB3 and JB6: $0.00037449 \text{ kg seed/ kg (wet) primary yield} * \text{kg primary yield}/0.18 \text{ kg DM} * 0.4 * 10^3 \text{ kg DM/ha} = 0.8322 \text{ kg seed/ha}$;

As a second step, the life cycle inventory for producing the seed must be established. For all seeds from cereals (barley and wheat), the same inventory data as used for the production of the grain can be used (Nemecek & Kägi, 2007), to which a transport and seed processing process (drying, cleaning) is added. This procedure is also used for maize, sugarbeet and ryegrass. For barley, wheat and maize, a 1:1 relation can be used, i.e. 1 kg grain from the main crop is needed to produce 1 kg seed. This can be translated in terms of ha needed of the main crop (per kg of seed) based on the corresponding yields (for maize, the conversion is done considering $12.05 * 10^3 \text{ kg DM/ha} * \text{kg primary yield}/0.31 \text{ kg DM} = 38870.97 \text{ kg/ha}$ for maize on JB3 and $11.93 * 10^3 \text{ kg DM/ha} * \text{kg primary yield}/0.31 \text{ kg DM} = 38483.87 \text{ kg/ha}$ for maize on JB6).

Two different transport distances are considered: from the seed producing farm to the processing centre and from the processing centre/regional storehouse and from there to the farm where the seeds will be sown. This consists of 30 km by lorry for the former and for the latter (Ecoinvent process "Transport, lorry >32t, EURO3/RER U", described in Spielmann *et al.*, 2007).

As a first proxy, the seed processing process is taken as in the Ecoinvent database. The seed processing is, in the Ecoinvent database, translated in terms of electricity consumption only. The drying energy required could alternatively have been estimated based on psychometric chart knowing the initial and final humidity of the grains. The electricity used is the marginal electricity for Denmark defined for Denmark when performing life cycle assessment (see for example Hamelin *et al.*, 2011).

For sugarbeet, based on Nemecek & Kägi (2007), the process for seed should be adjusted as compared to the production of the main crop in order to take into account, among others, the difference in yield. For sugarbeet seed, a yield of 20 hkg ha^{-1} is considered (Danish Ministry of Food, Agriculture and Fisheries, 2009). The yield for the main crop is, for both soils JB3 and JB6, 566 hkg ha^{-1} . Therefore, a quantity of 28.3 kg ($56600/2000 = 28.3$) sugar beet was taken as an input per kg seed produced (instead of 1:1 as for cereals). This results in $5 * 10^{-4} \text{ ha}$ of sugar beet needed per kg of seed ($28.3/566 * 10^2$).

For ryegrass seed, a yield of 12 hkg/ha is considered (Danish Ministry of Food, Agriculture and Fisheries, 2009). The yield of the main crop is, for soil JB3, $9.98 \text{ Mg DM ha}^{-1}$ (Table 2), and the DM content is 18.0% (Møller *et al.*, 2000). This means a wet yield of about 55444 kg ha^{-1} for ryegrass main crop on JB3 (and 49000 kg ha^{-1} on JB6). Therefore, for soil JB3, a quantity of 46.2 kg ($55444/1200 = 46.2$) ryegrass was taken as an input per kg seed produced. Using ryegrass DM yield for soil JB6 ($8.82 \text{ Mg DM ha}^{-1}$), a quantity of 40.83 kg ryegrass is

needed as an input per kg seed produced. This can be translated to 8.33×10^{-4} ha main crop needed per kg seed (JB3 and JB6).

Table S4 summarizes the life cycle inventory data to be used for seeds.

Table S4. Life cycle inventory for seeds

Output data	Unit	Input data ha of main crop	Other inputs	Comment
Barley seed	1 kg seed	JB3: 2.00×10^{-4} JB6: 1.75×10^{-4}	Transport Seed processing	Based on Ecoinvent process "Barley seed IP, at regional storehouse/CH U".
Wheat seed	1 kg seed	JB3: 1.52×10^{-4} JB6: 1.25×10^{-4}	Transport Seed processing	Based on Ecoinvent process "Wheat seed IP, at regional storehouse/CH U".
Sugar beet seed	1 kg seed	JB3 and JB6: 5.00×10^{-4}	Transport Seed processing	Based on Ecoinvent process "Sugar beet seed IP, at regional storehouse/CH U", adjusted for yield, see text.
Silage maize seed	1 kg seed	JB3: 2.57×10^{-5} JB6: 2.60×10^{-5}	Transport Seed processing	Based on Ecoinvent process "Maize seed IP, at regional storehouse/CH U".
Ryegrass seed	1 kg seed	JB3 and JB6: 8.33×10^{-4}	Transport Seed processing	Based on Ecoinvent process "Grass seed IP, at regional storehouse/CH U", adjusted for yield, see text.

For *Miscanthus*, the procedure differs, as cultivation of *Miscanthus* for rhizomes is performed differently than cultivation of *Miscanthus* for stem (Atkinson, 2009; Bullard and Metcalfe, 2001). First, a 3 year cycle is assumed for the production of rhizomes. This is identical as shown in Table S1, but in year 3, instead of harvesting (a part of) the stems, the rhizomes are harvested. In this process, rhizomes are lifted (in order to loosen the rhizomes mass), harvested and separated from soil (Atkinson, 2009). Atkinson (2009) suggests that field lifting of rhizomes is achieved using conventional rotary cultivator. This is also what Bullard and Metcalfe (2001) assumed. Rhizomes are then harvested and a potato planter is used to separate rhizomes from the soil, based on Atkinson (2009) as well as Bullard and Metcalfe (2001). Energy requirement for this specific process can be estimated as the same for standard potato grading operations (Bullard and Metcalfe, 2001).

The process for 1 ha rhizome production could therefore roughly be constituted of "Miscanthus production year 1, 1 ha", "Miscanthus production year 2, 1 ha", "Miscanthus production year 3, 1 ha, without stem

harvest”, “harvesting, by complete harvester, potatoes, 1 ha” (taken as a proxy for the rhizome harvesting), “potato planter, 1 ha” (separating rhizomes from the soil), and “tillage, rotary cultivator, 1 ha” (field lifting of rhizomes). Diesel consumption is adjusted in function of the soil type as in Dalgaard *et al.* (2001).

In a nutshell, for 1 ha rhizome, the following inputs are assumed:

- Herbicide application: (5.13 kg ha⁻¹ glyphosate (twice this dose), 3.4 kg ha⁻¹ dinitrianiiline compounds, 0.763 kg ha⁻¹ phenoxy-compounds, 2.2 kg ha⁻¹ nemzoic compounds, 0.02 kg ha⁻¹ [sulfonyl]urea-compounds, 0.015 kg ha⁻¹ “pesticide unspecified”) (same as received for *Miscanthus* in year 1, year 2 and year 3, as described in item 5 of this appendix)
- Mineral fertilizers: 15 plus 30 kg ha⁻¹ of nitrogen fertilizers, 25.4 plus 16.4 kg phosphorus fertilizer (as P₂O₅) and 70.8 kg plus 50.9 kg/ha potassium fertilizer (as K₂O) (same as for *Miscanthus*, year 2 plus year 3, described in item 6 of this appendix)
- Slurry fertilizers: same as *Miscanthus*, year 2 plus *Miscanthus*, year 3 (described in item 6 of this appendix)
- Planting of rhizomes
- Field emissions of carbon, nitrogen and other substances flows for year 2 and year 3
- Lifting rhizomes
- Separating rhizomes from the soil
- Harvesting rhizomes
- Transport of harvested rhizomes from field to farm (described in item 9 of this appendix)

Moreover, it is considered that 1 ha motherfield can deliver rhizomes for planting 5 ha (at 20000 rhizomes ha⁻¹).

For willow, 300 000 cutting ha⁻¹ are assumed in the cutting field. This means that 1 ha cutting delivers cuttings for 20 ha willow field (for which the planting density is 15 000 cuttings per ha). Field for cutting production are harvested annually, and the first harvest occurs on life cycle year 3 (second growth year). One ha cutting therefore comprises all the processes included in willow production for life cycle year 1, 2 and 3 (without herbicide application for that year). Transport of the cuttings to the farm is included, assuming a weight of 100 g per cutting.

The methodology described above (for all crops) assumes that the seeds, cuttings and rhizomes are produced in Denmark, which may not be true in practice, especially for e.g. maize. Best practices would require to build the inventory based on the origin of the marginal seeds. Another simplification was made in the modelling. This relates with the crops involving the possibility of a secondary harvest (spring barley, winter wheat and sugar beet). When a crop has its secondary yield (e.g. straw) incorporated, it is considered that the seed to produce it comes from a plantation where the straw is harvested. This is not likely to mean much when performing life cycle assessments, it is simply mention for transparency purposes.

4. Liming

Lime is used on agricultural soils in order to maintain a soil pH ensuring optimal crop nitrogen uptake. Lime is therefore used for soils with low pH, so it is not a crop-related input but rather a site-related input. Different type of lime may be used, but the most used in Denmark is calcium carbonate (CaCO_3) (Gyldenkærne *et al.*, 2005). According to Dankalk (Dankalk, 2011), the primary material for the production of agricultural lime in Denmark is whether white chalk from North Jutland or calcined limestone. For such application of lime obtained directly from the ground, Nemecek & Kägi (2007) recommend to use the Ecoinvent process “limestone, milled, packed, at plant CH” from Althaus *et al.* (2007). Lime could also emerge as a by-product of the manufacture of other products, e.g. sugar. Yet, this type of lime would not react to a change in demand for the crop considered in this study, and therefore cannot be the marginal. In this study, the process “limestone, milled, packed, at plant CH” from Althaus *et al.* (2007) is used.

Based on the Danish National Inventory Report for the United Nations Framework Convention on Climate Change (UNFCCC) (Nielsen *et al.*, 2009), the total amount of lime used on Danish agricultural soils for the last 5 years of data (2003 to 2007) is 2 237 000 ton of pure calcium carbonate (CaCO_3) while the amount of agricultural area for this same period was, on average, 2 676 703 ha. Using these figures, a rough estimate can be made for a 5-years lime consumption of 0.8357 ton per hectare. For this inventory, it is therefore considered that $0.8357 \text{ ton ha}^{-1}$ of CaCO_3 is applied per application, i.e. once every 5 years for annuals and once every 20 years for perennials.

It is considered that lime is applied with the same spreader as mineral fertilizers, i.e. they are applied simultaneously. Therefore, the Ecoinvent process “Fertilising, by broadcaster, CH/U” is used for the modelling. This process is adjusted to consider the specific diesel consumption for lime spreading. In this study, the value used is based on Dalgaard *et al.* (2001): $1.5 \text{ l diesel ha}^{-1} \text{ y}^{-1}$ (value uncorrected for soil type). This means that the diesel consumption is $1.5 \text{ l diesel ha}^{-1} \text{ y}^{-1}$ for soil JB6 (correction factor of 1 for sandy loam) and $1.35 \text{ l diesel ha}^{-1} \text{ y}^{-1}$ for soil JB3 (correction factor of 0.9 for sandy soils). The process used from the Ecoinvent database estimate many of the emissions to air based on the diesel consumed. These were corrected accordingly.

5. Sowing/planting

The process used for sowing is the Ecoinvent process “Sowing/CH U” (for all crops except willow and *Miscanthus*), adjusted with the diesel consumption presented in Dalgaard *et al.* (2001), i.e. 2.7 l ha^{-1} (soil JB3) and 3.0 l ha^{-1} (soil JB6). The emissions that are function of the amount of diesel burned are also adjusted accordingly.

For *Miscanthus*, planting may be done using a manure spreader followed by cultivation and rolling but this is not likely to represent future practices as it results in rather unpredictable plant spacing and establishment rate (DEFRA, 2007; Heaton *et al.*, 2004). A potato planter can also be adapted and used to plant the rhizomes, but

the optimal option consists to use a specialized *Miscanthus* planter (Heaton *et al.*, 2004). For willow, planting may be performed by a step planter.

For this study, the most important parameter to consider regarding sowing operations is the associated consumption of diesel related to the planting. Therefore, for these two crops, the Ecoinvent process “Planting, CH/U” has been used, which involve a diesel consumption of 20 l ha⁻¹. This consumption has been adjusted, for willow, based on Heller *et al.* (2003), whom used an empirical formula to estimate the diesel consumption, having as inputs the maximal available PTO power (which they estimated at 78 kW) and the total power required for the operation (which they estimated at 52 kW). Based on this, and on an operating rate of 2.5 h ha⁻¹ (Heller *et al.*, 2003) the diesel consumed for planting willow is estimated at 56.6 l ha⁻¹. As a proxy, this value will also be used for *Miscanthus*. In both cases, values are adjusted in function of the soil type, based on Dalgaard *et al.* (2001).

6. Plant protection

For each crop, application of pesticides products (herbicides, insecticides and fungicides) is an integral part of plant protection operations. Table S5 presents the pesticides types and the amount of each to be applied annually for annual crops and ryegrass. For ryegrass, values are integrated over the lifecycle and expressed annually. Sugarbeet data are from the “Dansk Landbrugs Grovvarereselskab” (DLG, 2009). This crop requires important chemical control, as compare to other annuals crops. All other data are from Henriksen *et al.* (submitted).

For each pesticides type, it is considered that the same amount is applied on both soil types. The inventory data for the production of these pesticides are taken from the Ecoinvent database (Nemecek & Kägi, 2007). The data from Nemecek & Kägi (2007) are, however, based on Green (1987), which are rather old data. These data only consider energy inputs. According to the authors, these data are nevertheless reliable, and can be apply for the European context (the data from Green, 1987, were derived for US plants whose patents expired, but they are the results of simulation models, not direct measurements), through they warn that they should not be used for anything else than for agricultural-related LCA.

Nemecek & Kägi (2007) argue that given the importance of patents in the pesticides industry, it is very hard to get recent data. The authors also highlight that the impact of pesticides is generally small in agricultural LCA, but emphasize that the toxicity can be significant.

In Nemecek & Kägi (2007), each pesticide is classified in classes, and the classes corresponding to the pesticides used in this study are presented in Table S5. Conformingly to the methodology suggested in Nemecek & Kägi (2007), pesticides not listed in the categories made by the authors are assigned to the general category “pesticides, unspecified”. Correspondence between trade name and compound were obtained through the pesticide properties database (PPDB) (Green, 2009). In this study, European data are used (RER), but adjusted with the marginal electricity defined for Denmark when performing life cycle assessment (see e.g. Hamelin *et al.*, 2011).

Table S5. Application of pesticides for annual crops and ryegrass, for both soil and climate types

Pesticide name	Pesticide class (Nemecek & Kägi, 2007), obtained from Green (2009)	Type*	Spring barley	Spring barley & Catch crop	Winter wheat	Maize silage	Sugar beet [†]	Ryegrass
Fluoroxypir	Phenoxy compounds	H			0.00350	0.0360		0.080
Foramsulfuron	Pesticide, unspecified	H				0.0300		
Pendimethalin	Dinitroaniline compounds	H				0.5000		
Idosulfuron	[sulfonyl]urea compounds [‡]	H				0.0133		
Bentazon	Benzo[thia]diazole compounds	H				0.2160		
Clopyralid	Benzoic compounds	H						0.040
MCPA	MCPA	H						0.400
Tribenuron-methyl	Triazine compounds	H	0.00375	0.00375				
Ioxynil	Nitrile compounds	H	0.05000	0.05000				
Pyrethrostrobin	Pyretroid compounds [‡]	F	0.06250	0.06250	0.02400			
Diflufenican	Diphenylether compounds	H			0.02000			
Bromoxynil	Nitrile compounds	H	0.05000	0.05000	0.02400			
Glyphosate	Glyphosate	H			0.72000			
Lamda-cyhalothrin	Pyretroid compounds	I	0.00375	0.00375	0.00375			
Prosulfocarb	[thio]carbamate compounds	H			0.80000			
Boscalid	Pesticide, unspecified	F			0.17475			
Florasulam	Pesticide, unspecified	H			0.04000			
Tebuconazole	Cyclic N compounds	F			0.06250			0.125
Epoxiconazole	Cyclic N compounds	F			0.14400			
Metamitron	Triazine compounds	H					2.45	
Phenmedipham	[thio]carbamate compounds	H					2.91	
Ethofumesate	Pesticide, unspecified	H					0.09	
Penetrating oil (Penol 33 E)	Pesticide, unspecified	oil					0.90	

* H: herbicide; F: fungicide; I: insecticide.

[†] Conversions in kg ha⁻¹ were performed based on the following densities: Metamitron: 1.225 g ml⁻¹; Phenmedipham: 0.97 g ml⁻¹; Ethofumesate: 1.3 g ml⁻¹ and Penol 33E: 0.9 g ml⁻¹.

[‡] Deduced, as this compound does not figure among the substances specified by Nemecek & Kägi (2007).

Miscanthus, in the establishment phase, is particularly vulnerable to competition with weeds. The year before planting (year 1), a systemic herbicide (glyphosate) is therefore applied to prevent important yield decreases due to competition with weeds. A dose of 3 l ha⁻¹ is considered, based on a personal communication with Jens Bonderup Kjeldsen, Aarhus University (Kjeldsen, 2010). After planting (year 2), a pre-emergence weed killer (pendimethalin, 4 l ha⁻¹) is applied. During the emergence (year 2), specific herbicides may be used depending on the weed present. In this study, based on a personal communication with Jens Bonderup Kjeldsen, Aarhus University (Kjeldsen, 2010) and a fact sheet from Irish research (Finnan and Caslin, 2008), it is considered that fluroxypyr (0.7 l ha⁻¹) clopyralid (1.25 l ha⁻¹), Metsulfuron-methyl (20 g ha⁻¹) and Thifensulfurol-methyl (15 g ha⁻¹) are applied. The third year, glyphosate is applied again, late April or early May where grass weeds are present (3 l ha⁻¹). After that, it is considered that the crop will surpass weed growth and therefore chemical

control is no longer required (Finnan and Caslin, 2008). Nevertheless, a treatment every 4th year after the first harvest is considered (glyphosate pre emergence, 3 l ha⁻¹: year 8, year 12 and year 16), in order to clean up for potential problematic weed species. This is based on a personal communication with Jens Bonderup Kjeldsen, Aarhus University (Kjeldsen, 2010). Chemical weed treatments for *Miscanthus*, over its life cycle, is summarised in Table S6.

Table S6. Pesticides applied to *Miscanthus* (autumn and spring harvest) over the 20 year life cycle, for both soil and climate types

Pesticide	Type*	Pesticide class (Nemecek & Kägi, 2007)	Amount over lifecycle (kg ha ⁻¹)	Comment
Glyphosate	H	Glyphosate	25.65	Before planting (year 1) and the 3 rd year. Also on year 8, 12 and 16. Amount of 3 l ha ⁻¹ per application. Density of 1.71 g ml ⁻¹ used for conversion.
Pendimethalin	H	Dinitroaniline compounds	3.4	Applied on year 2. Density of 0.85 g ml ⁻¹ used for conversion.
Fluroxypyr	H	Phenoxy compounds	0.763	Applied on year 2. Density of 1.09 g ml ⁻¹ used for conversion.
Clopyralid	H	Benzoic compounds	2.20	Applied on year 2. Density of 1.76 g ml ⁻¹ used for conversion.
Metsulfuron- methyl	H	[sulfonyl]urea compounds	0.02	Applied on year 2.
Thifensulforol- methyl	H	Pesticides, unspecified	0.015	Applied on year 2.

* H: herbicide; F: fungicide; I: insecticide.

As *Miscanthus*, willow is particularly vulnerable to competition with weed during the establishment. Once the plantation is established, a high amount of weed is tolerated. The year before planting (year 1), a systemic herbicide is applied to control perennial weeds. A dose of 3 l ha⁻¹ is considered, based on a personal communication with Jens Bonderup Kjeldsen, DJF, Aarhus University (Kjeldsen, 2010). Shortly after planting (year 2), pendimethalin (4 l ha⁻¹) is applied to ensure a good, fine seedbed. During the establishment (year 3), various herbicides may be applied, depending on the amount of weed. In this study, the following are considered: fluazifop-p-butyl (2 l ha⁻¹) and clopyralid (1.25 l ha⁻¹) (based on a personal communication with Jens Bonderup Kjeldsen, DJF, Aarhus University, (Kjeldsen, 2010), and a factsheet for chemical weed control of willow in Ireland, (Finnan and Caslin, 2008a)). After the first harvest (year 6), glyphosate is applied to ensure the plantation is kept weed free (3 l ha⁻¹). This is then performed every other harvest (harvest then occur every 3 years, meaning next applications will take place on year 12 and 18). Once canopy closure occurs (a few months after the harvest), the coppice controls its own weeds due to reduced light levels reaching the ground surface (Finnan and Caslin, 2008a). At maturity, the underlying vegetation does not cause any detrimental effects to the plantation and may even contribute to prevent pest species as it provides an habitat for predators of some pests. Therefore, no pesticides are applied during this period. Chemical plant protection is to some extent a dynamic activity, and more or less applications may occur depending on the actual conditions on the field for a particular year. It is nevertheless judged that the doses used in this study are representative

of a how a “typical” Danish willow plantation would be managed. The overall pesticide application for willow lifecycle is summarised in Table S7.

Table S7. Pesticides applied to willow over its 21 years lifecycle, for both soil and climate types

Pesticide	Type*	Pesticide class (Nemecek & Kägi, 2007)	Amount over lifecycle (kg ha ⁻¹)	Comment
Glyphosate	H	Glyphosate	20.52	4 times 3 l ha ⁻¹ : (year 1, 6, 12 and 18). Density of 1.71 g ml ⁻¹ used for conversion.
Pendimethalin	H	Dinitroaniline compounds	3.4	Applied on year 2. Density of 0.85 g ml ⁻¹ used for conversion.
Fluazifop-p-butyl	H	Phenoxy compounds	2.44	Applied on year 3. Density of 1.22 g ml ⁻¹ used for conversion.
Clopyralid	H	Benzoic compounds	2.20	Applied on year 3. Density of 1.76 g ml ⁻¹ used for conversion.

*H: herbicide; F: fungicide; I: insecticide.

Application of pesticides is carried out by a field sprayer with an 800 l carrying capacity, based on Nemecek & Kägi (2007) (process “Application of plant protection products, by field spreader/CH U”). This process is adjusted for diesel consumption based on the norm data presented by Dalgaard *et al.* (2001), i.e. a consumption of 1.35 l ha⁻¹ is considered on soil JB3 and of 1.5 l ha⁻¹ on soil JB6. The emissions dependent upon diesel burning have been adjusted consequently.

7. Fertilization

As described in the manuscript, half on the nitrogen (N) requirements are provided by animal slurry (50 % fattening pig and 50 % dairy cattle) and half through mineral fertilizers (calcium ammonium nitrate, with a sensitivity analysis with urea). For phosphorus (P) and potassium (K), the balance not fulfilled by animal slurry is provided by mineral fertilizers (diammonium phosphate and potassium chloride, respectively).

The data for both urea and calcium ammonium nitrate production are taken from the Ecoinvent processes “Calcium ammonium nitrate, as N, at regional storehouse/RER U” and “Urea, as N, at regional storehouse/RER U”. These represent European data, though the marginal producing technology for urea may in practice not be European. European data are nevertheless used as a best proxy.

The production of calcium ammonium nitrate requires nitric acid, and this is included in the abovementioned Ecoinvent process. However, the data for nitric acid production in the Ecoinvent database are from 1997, and there are reasons to believe that the process should be adapted in order to represent more up-to-date plant conditions. For example, Börjesson and Tufvesson (2011) mention that approximately half of the nitrogen fertilizer plants in Western Europe have installed catalytic cleaning equipment (reducing N₂O by some 80 %), and they further add that within the next few years, all plants are expected to have such technology. The authors report an average emission of 3 g N₂O per kg N produced for these plants. Using, based on the Ecoinvent data, an amount of 2.25 kg acid nitric per kg of calcium ammonium nitrate (as N) produced, this represents an emission of 0.0013 kg N₂O per kg nitric acid produced. The actual Ecoinvent process includes

0.00839 kg N₂O per kg nitric acid, which is about 6 times higher. The EU BREF document on the manufacture of large volume inorganic chemicals (EC, 2007) indicates emission values between these 2 extremes, for plants across Europe. In fact, an average of 42 plants data (EC 2007, Table 3.7) presented in the BREF lead to an average of 0.0062 kg N₂O per kg nitric acid. The BREF also presents values for plants applying catalytic N₂O decomposition in the oxidation reactor (EC 2007, Table 3.12), and an average of these result in a value of 0.00248 kg N₂O per kg nitric acid, which is still about twice as high as the value presented by Börjesson and Tufvesson (2011). The best available techniques (BAT) level stated in the BREF is 0.00012-0.00185 kg N₂O per kg nitric acid for existing plants (with a split view saying the upper limit should be 0.0025). For new plants, it is 0.00012 to 0.0006 kg N₂O per kg nitric acid.

In this study, a value of 0.0062 kg N₂O per kg nitric acid has been used, which represents the average of the plants reported in the BREF (EC, 2007). This may be seen as a higher end-of-interval value as it is well above BAT emission levels, but it is still about 26 % lower than the original value displayed by the Ecoinvent database.

The application of mineral fertilisers are included by the Ecoinvent process "Fertilising, by broadcaster". Diesel consumption is adjusted based on the norm values presented by Dalgaard *et al.* (2001), which corresponds to 1.8 L ha⁻¹ on soil JB3 and 2 L ha⁻¹ on soil JB6. The emissions dependent upon diesel burning are adjusted in consequence.

For both fattening pig and dairy cow, the slurry composition was determined based on the exact same methodology and assumptions as presented in the Danish study of Wesnæs *et al.* (2009). In their study, Wesnæs *et al.* (2009) established a reference slurry composition for both fattening pig and dairy cow slurry in order to carry out a life cycle assessment of slurry management technologies in Denmark. The methodology presented in Wesnæs *et al.* (2009) takes for basis the Danish normative system for assessing manure composition, for which Poulsen *et al.* (2001) established the technical background report. As in Wesnæs *et al.* (2009), the values for DM (ex-storage), N, P and K (ex-animal) used are based on annually updated Danish manure standards. In the present case, however, the latest updated values (at the moment of carrying out the study) will be used, i.e. those in Poulsen (2009) (Wesnæs *et al.* (2009) used the 2008 data). This is why the N, P and K values differ slightly from those presented by Wesnæs *et al.* (2009). However, since the DM (ex-storage) has the same value in manure standards 2008 and 2009, all other values are the same (since these values are all dependent upon DM). These values assumed that the slurry is stored in a concrete tank (for pig, cut straw is added as a floating layer; for cows, it is assumed that a natural crust is forming). Concerns related to the use of the data from the manure standards are expressed in Wesnæs *et al.* (2009), since these data do not consider water addition in the housing system, which results in higher ex-storage concentrations than those found in practice as the slurry is not diluted. These data are nevertheless used as they are considered as the "Danish standard data" for the majority of Danish studies about slurry (Wesnæs *et al.*, 2009).

As discussed in Wesnæs *et al.* (2009) as well as in Hamelin *et al.* (2011), the slurry composition is the basis for assessing the nutrient flow from the slurry in the environment. Yet, the slurry composition may be highly variable from one farm to another, depending, among others, on the diet, the slurry management, the housing system, the in-house environmental conditions, the storage time, etc. Through the slurry composition used in

this study is based on best available Danish data, it does not pretend to represent the “average Danish slurry” for both fattening pigs and dairy cows, so any use of the data presented in this study must be done in the light of this consideration.

Tables S8 and S9 present the slurry composition used for this study, for fattening pigs and dairy cows, respectively. The values of interest for the present study are the values ex-storage (i.e. before application), but the values for slurry ex-housing and post-animal are given for information. The number of digits should not be seen as an indication of precision of the values; many digits were conserved as rounding data at this stage is likely to result in inconsistencies in the different mass balances to be carried out in later stages.

Table S8. Composition of the pig slurry to be used as an organic fertilizer

	Slurry ex-animal	Slurry ex-housing	Slurry ex-storage	Source and assumptions
Total mass (ton)	1	1	1	
Total N (kg)	6.565	5.449	4.765	N ex-animal from Poulsen (2009) (3.02 kg / 0.46 ton). Values ex-housing and ex-storage obtained from mass balances. Losses considered (during housing and during storage): NH ₃ , N ₂ O, N ₂ , NO. See Table S10 for details about N losses. Ex-storage values adjusted considering water addition of 86 kg during the storage.
P (kg)	1.087	1.087	1.001	P ex-animal from Poulsen (2009) (0.50 kg / 0.46 ton). No losses considered. Ex-storage values adjusted considering water addition of 86 kg during the storage.
K (kg)	2.848	2.848	2.622	K ex-animal from Poulsen (2009) (1.31 kg / 0.46 ton). No losses considered. Ex-storage values adjusted considering water addition of 86 kg during the storage.
DM (kg)	77.481	69.733	61.000	DM ex-storage from Poulsen (2009). Losses during storage: 5 % of the ex-housing values. Losses during housing: 10 % of the ex-animal value. Water addition of 86 kg during storage is taken into account.
Ash (kg)	13.249	13.249	12.200	Ash ex-storage = 20 % of DM ex-storage. No losses considered. Water addition of 86 kg during storage is taken into account.
VS (kg)	64.232	56.483	48.800	VS ex-storage = 80 % of DM ex-storage. Losses considered during storage and housing (absolute values) are the same as for DM (all DM loss was VS). Water addition of 86 kg during storage is taken into account.
C (kg)	37.113	33.402	29.219	C ex-storage = 47.9 % of DM ex-storage, based on ratio C:DM obtained by Knudsen & Birkmose (2005). Losses assumed to follow the same pattern as DM: losses during storage of 5 % of the ex-housing value and losses during housing of 10 % of the ex-animal value. Water addition of 86 kg during storage is taken into account.
Cu (g)	30.009	30.009	27.633	Cu ex-storage = 0.0453 % of DM ex-storage, based on ratio Cu:DM obtained by Knudsen & Birkmose (2005). No losses considered. Water addition of 86 kg during storage is taken into account.
Zn (g)	89.432	89.432	82.350	Zn ex-storage = 0.135 % of DM ex-storage, based on ratio Zn:DM obtained by Knudsen & Birkmose (2005). No losses considered. Water addition of 86 kg during storage is taken into account.
Density (kg m ⁻³)	1053	1053	1053	From literature references for slurry density (Sherlock <i>et al.</i> , 2002; Sanchez & Gonzalez, 2004). To be seen as an indicative value rather than as an exact value.
pH			7.8	Based on Sommer & Husted (1995).

Table S9. Composition of the dairy cattle slurry to be used as an organic fertilizer

	Slurry ex-animal	Slurry ex-housing	Slurry ex-storage	Source and assumptions
Total mass (ton)	1	1	1	
Total N (kg)	6.892	6.363	5.807	N ex-animal from Poulsen (2009) (140.6 kg / 20.4 ton). Values ex-housing and ex-storage obtained from mass balances. Losses considered (during housing and during storage): NH ₃ , N ₂ O, N ₂ , NO (Table S10). Addition of N through straw added in the stable are accounted for (Table S11). Ex-storage values adjusted considering water addition of 44 kg during the storage.
P (kg)	1.020	1.032	0.989	P ex-animal from Poulsen (2009) (20.8 kg / 20.4 ton). Addition of P through straw added in the stable are accounted for (see Table S11). No losses considered. Ex-storage values adjusted considering water addition of 44 kg during the storage.
K (kg)	5.819	6.088	5.831	K ex-animal from Poulsen (2009) (118.7 kg / 20.4 ton). Addition of K through straw added in the stable accounted for (see Table S11). No losses considered. Ex-storage values adjusted considering water addition of 44 kg during the storage
DM (kg)	125.768	113.192	103.000	DM ex-storage from Poulsen (2009). Losses during storage: 5 % of the ex-housing values. Losses during housing: 10 % of the ex-animal value. Water addition of 44 kg during storage is taken into account.
Ash (kg)	21.506	21.506	20.600	Ash ex-storage = 20 % of DM ex-storage. No losses considered. Water addition of 44 kg during storage is taken into account.
VS (kg)	104.262	91.685	82.400	VS ex-storage = 80 % of DM ex-storage. Losses considered during storage and housing (absolute values) are the same as for DM (all DM loss was VS). Water addition of 44 kg during storage is taken into account.
C (kg)	55.212	49.691	45.217	C ex-storage = 43.9 % of DM ex-storage, based on ratio C:DM obtained by Knudsen & Birkmose (2005). Losses assumed to follow the same pattern as DM: losses during storage of 5 % of the ex-housing value and losses during housing of 10 % of the ex-animal value. Water addition of 44 kg during storage is taken into account.
Cu (g)	12.151	12.151	11.639	Cu ex-storage = 0.0113 % of DM ex-storage, based on ratio Cu:DM obtained by Knudsen & Birkmose (2005). No losses considered. Water addition of 44 kg during storage is taken into account.
Zn (g)	23.334	23.334	22.351	Zn ex-storage = 0.0217 % of DM ex-storage, based on ratio Zn:DM obtained by Knudsen & Birkmose (2005). No losses considered. Water addition of 44 kg during storage is taken into account.
Density (kg/m ³)	1053	1053	1053	From literature references for slurry density (Sherlock <i>et al.</i> , 2002; Sanchez & Gonzalez, 2004). To be seen as an indicative value rather than as an exact value.
pH			7.8	Based on Sommer & Husted (1995).

Table S10. Assumptions for N losses in the establishment of the slurry composition

Losses	Pig slurry	Dairy cow slurry
Losses in-house (kg)		
NH ₃ -N	16 % of N ex-animal (Poulsen <i>et al.</i> , 2001)	8 % of N ex-animal (Poulsen <i>et al.</i> , 2001)
N ₂ O-N	0.002 kg N ₂ O-N per kg N ex-animal (IPCC, 2006)	0.002 kg N ₂ O-N per kg N ex-animal (IPCC, 2006)
N ₂ -N	Assumption that N ₂ -N = N ₂ O-N * 3 (based on data from Dämmgen and Hutchings, 2008)	Assumption that N ₂ -N = N ₂ O-N * 3 (based on data from Dämmgen & Hutchings, 2008)
NO-N	Assumption that N ₂ -N = N ₂ O-N * 1 (based on data from Dämmgen & Hutchings, 2008)	Assumption that N ₂ -N = N ₂ O-N * 1 (based on data from Dämmgen & Hutchings, 2008)
Losses during storage (kg)		
NH ₃ -N	2 % of N ex-housing (Poulsen <i>et al.</i> , 2001), the N ex-housing being estimated according to Poulsen <i>et al.</i> (2001), i.e. : N ex-animal – NH ₃ -N losses in-house (and not accounting for other losses).	2 % of N ex-housing (Poulsen <i>et al.</i> , 2001), the N ex-housing being estimated according to Poulsen <i>et al.</i> (2001), i.e. : N ex-animal – NH ₃ -N losses in-house (and not accounting for other losses) + N from straw addition.
N ₂ O-N	0.005 kg N ₂ O-N per kg N ex-animal (IPCC, 2006)	0.005 kg N ₂ O-N per kg N ex-animal (IPCC, 2006)
N ₂ -N	Assumption that N ₂ -N = N ₂ O-N * 3 (based on data from Dämmgen & Hutchings, 2008)	Assumption that N ₂ -N = N ₂ O-N * 3 (based on data from Dämmgen & Hutchings, 2008)
NO-N	Assumption that N ₂ -N = N ₂ O-N * 1 (based on data from Dämmgen & Hutchings, 2008)	Assumption that N ₂ -N = N ₂ O-N * 1 (based on data from Dämmgen & Hutchings, 2008)

Table S11. Assumptions for N, P and K balance through straw addition in the stable, for dairy cows

Parameter	Value
kg straw animal ⁻¹ day ⁻¹	1.2 (Poulsen <i>et al.</i> , 2001)
Straw DM (%)	85 (Poulsen <i>et al.</i> , 2001)
kg N kg ⁻¹ DM	0.005 (Poulsen <i>et al.</i> , 2001)
kg P kg ⁻¹ DM	0.00068 (Poulsen <i>et al.</i> , 2001)
kg K kg ⁻¹ DM	0.01475 (Poulsen <i>et al.</i> , 2001)
kg slurry animal ⁻¹	20 400 (Poulsen, 2009)
Addition of N, P and K from straw per ton slurry ex-animal	0.0913 kg N; 0.0124 kg P; 0.269 kg K (calculated based on the above parameters)

Slurry spreading is performed by trail hose application tanker, as this is judged representative of Danish conditions (Wesnæs *et al.* 2009). This process is modelled by the Ecoinvent process “Slurry spreading, by vacuum tanker”. The process includes the diesel consumed for slurry application, construction of the tractor,

the slurry tanker and a shed. Emissions from the diesel consumption by the tractor are adjusted based on the norm values presented by Dalgaard *et al.* (2001), which corresponds to 0.54 l t⁻¹ slurry on soil JB3 and 0.60 l t⁻¹ slurry on soil JB6 (this includes loading). The emissions caused by burning diesel are adjusted in consequence.

The annual fertilization needs for each of the selected crop is based on the fertilization guidelines issued by the Danish Ministry of Food, Agriculture and Fisheries (Danish Ministry of Food, Agriculture and Fisheries, 2009), except for *Miscanthus*. These are what the farmers use to make their fertilisation plan.

Table S12 presents the needs in N, P and K for each of the selected crop, on both a sandy soil and a sandy loam soil.

Table S12. Fertilization needs for all selected crops on sandy and sandy loam soils, for both climate types

Crop	Sandy soil			Sandy loam soil		
	N	P	K	N	P	K
	kg ha ⁻¹					
Spring barley	126	22	45	114	22	45
Spring barley & Catch crop *	109	22	45	97	22	45
Winter wheat	166	22	66	161	22	66
Willow	120	15	50	120	15	50
<i>Miscanthus</i> (autumn) ^{††}	90	15	75	90	15	75
<i>Miscanthus</i> (spring) ^{††}	60	15	75	60	15	75
Maize silage	162	44	135	139	44	135
Sugar beet	123	43	150	102	43	150
Ryegrass	342	36	239	325	36	239

*Catch crops are assumed to reduce the N norm by 17 kg N ha⁻¹.

[†]For N, values come from Olesen *et al.* (2001).

^{††}For N, these are the values for year 4 to 20 (established *Miscanthus*). No N is applied in year 1, 30 kg ha⁻¹ is applied on year 2 and 60 kg ha⁻¹ is applied on year 3, for both spring and autumn harvest.

8. Irrigation

Irrigation is applied only when crops are grown on sandy soils, except for willow and *Miscanthus*, for which it is considered that irrigation is not economically profitable under Danish conditions. Irrigation has been considered through the Ecoinvent process “Irrigating/ha/CH U”, described in Nemecek & Kägi (2007). Adjustments regarding the electricity source were made, through, as the process is run in Denmark.

This process considers that irrigation is performed with a mobile sprinkler system, with the irrigation water coming from surface water, with an annual water sprayed of 1200 m³ per ha (4 times 300 m³ water). This includes the fix installed pump (30 m³ h⁻¹, 7 to 8 bar with a 22 kW engine), the polyethylene main water pipe, the excavation (for the main pipe) as well as the tractor needed to install the equipment on the field. Also included are the hydrant (valve assembly unit), the mobile turbine-driven irrigation automaton and the PVC

water hose to link the automaton to the hydrant. The energy used is also included, considering 880 kWh ha^{-1} ($22 \text{ kW} * 1200 \text{ m}^3/\text{ha} * \text{h}/30 \text{ m}^3 = 880 \text{ kWh/ha}$).

9. Harvest

The environmental impacts from harvesting operations are modelled through the Ecoinvent process “Combine harvesting/CH U” for spring barley and winter wheat. This process is adjusted to consider the specific diesel consumption involved in this study. In this study, the value used is based on Dalgaard *et al.* (2001): $14 \text{ l diesel ha}^{-1} \text{ y}^{-1}$ (value uncorrected for soil type). This means the diesel consumption is $14 \text{ l diesel ha}^{-1} \text{ y}^{-1}$ for soil JB6 (correction factor of 1 for sandy loam) and $12.6 \text{ l diesel ha}^{-1} \text{ y}^{-1}$ for soil JB3 (correction factor of 0.9 for sandy soils). The process used from the Ecoinvent database estimates many of the emissions to air based on the diesel consumed. These were corrected accordingly.

Harvesting of maize is modelled through the Ecoinvent process “Chopping, maize/CH U”, with a diesel consumption of 35 l ha y^{-1} for soil JB3 and of 39 l ha y^{-1} for soil JB6. This is based on Dalgaard *et al.* (2001) and adjusted for soil type. Since the values in Dalgaard *et al.* (2001) for “chopping” are expressed in l t^{-1} , the primary yield for maize (average for soil JB3 and JB6) was used for the conversion in l ha^{-1} , considering a DM content of 31.0 %, based on Møller *et al.* (2000) (value for “medium content of digestible carbohydrate”). Loading the maize is included through the Ecoinvent process “Fodder loading, by self-loading trailer/CH U”. No changes are performed for the diesel consumption (0.106 kg m^{-3} fodder). However, the process is expressed per m^3 , so it is needed to convert it per ha. The Ecoinvent process “silage maize IP, at farm” assumes 0.004 m^3 fodder loading per kg fresh weight (FW) of silage maize. Based on this, and on the yield data for soils JB3 and JB6, this would correspond to $155.48 \text{ m}^3 \text{ ha}^{-1}$ on JB3 and $153.94 \text{ m}^3 \text{ ha}^{-1}$ on JB6. (Example for JB3: $12.05 \text{ t DM/ha} * \text{kg FW}/0.31 \text{ kg DM} * 0.004 \text{ m}^3 \text{ fodder loading/kg FW} * 1000 \text{ kg/t} = 155.48 \text{ m}^3 \text{ ha}^{-1}$).

For sugar beets, the process used is the Ecoinvent process “Harvesting, by complete harvester, beets/CH U”, with a diesel consumption adjusted to 27 l ha^{-1} (soil JB6) and 24.3 L ha^{-1} (soil JB3), based on Dalgaard *et al.* (2001). This includes cutting the beet tops.

Due to its high moisture content, *Miscanthus* harvested in autumn is not suited for being harvested by big baling system (Kristensen, 2003). Therefore, it is considered that autumn harvest *Miscanthus* is harvested by exact chopper, so it can be used directly in a combustion plant. The Ecoinvent process “Chopping, maize/CH U”, is used as a best proxy for modelling the impacts related to harvesting. The diesel consumption is adjusted based on a fuel use of 52 l h^{-1} (Smeets *et al.*, 2009: two 75 kw tractors consuming 17 l h^{-1} of fuel each and 1 chopper consuming 18 l h^{-1}) and a net capacity of 11.4 t DM h^{-1} (Kristensen, 2003: average of the three values presented). Based on yields for autumn harvested *Miscanthus*, the diesel consumption considered in this study is therefore:

- JB6, wet and dry climate: 69.6 L ha^{-1} (year 4-20), 0 (year 2: no harvest), 25.0 L ha^{-1} (year 3);
- JB3, wet climate: 62.6 L ha^{-1} (year 4-20), 0 (year 2: no harvest), 22.5 L ha^{-1} (year 3);
- JB3, dry climate: 53.2 L ha^{-1} (year 4-20), 0 (year 2: no harvest), 19.2 L ha^{-1} (year 3)

(Example for wet climate, JB6, year 4-20: $15.25 \text{ t DM/ha} * \text{h}/11.4 \text{ t DM} * 52 \text{ l/h} * 1.0$ (correction factor JB6) = 69.6 L ha^{-1}).

These numbers include the correction for soil type.

Miscanthus harvested in spring is mowed and baled by a big baler. The bale density is between 140 to 170 kg m^{-3} and the power requirement is about 20.6 kW (at PTO) (Kristensen, 2003). The diesel consumption is based on a fuel use of 52.5 l h^{-1} (Smeets *et al.*, 2009: three 75 kW tractors consuming 17 l h^{-1} of fuel each and 1 big baler consuming 1.5 l h^{-1}) and a net capacity of 13.2 t DM h^{-1} (Kristensen, 2003: average of the three values presented). Based on yields for spring harvest *Miscanthus*, the diesel consumption is therefore:

- JB6, wet and dry climate: 39.8 L ha^{-1} (year 4-20), 0 (year 2: no harvest), 14.3 L ha^{-1} (year 3)
- JB3, wet climate: 35.8 L ha^{-1} (year 4-20), 0 (year 2, no harvest), 12.9 L ha^{-1} (year 3)
- JB3, dry climate: 30.4 L ha^{-1} (year 4-20), 0 (year 2, no harvest), 11.0 L ha^{-1} (year 3)

In order to model this process, the process “Baling/ CH U” from the Ecoinvent database has been used. The environmental flows involved by this process are, however, expressed per bale (instead of per ha as in this study), considering 160 kg (fresh weight) per bale (Nemecek & Kägi, 2007). To express the flows per ha, the primary yield as well as the moisture content are needed. Moisture content are taken from Kristensen (2003), i.e. $0.85 \text{ kg DM kg}^{-1}$ fresh weight for spring harvest *Miscanthus* (JB3) and $0.904 \text{ kg DM kg}^{-1}$ fresh weight for spring harvest *Miscanthus* (JB6). The flows inventoried in the Ecoinvent process should therefore be multiplied by:

- JB3, wet climate: $73.5 \text{ bale ha}^{-1}$ (year 4-20) and $26.5 \text{ bale ha}^{-1}$ (year 3)
- JB3, dry climate: $62.5 \text{ bale ha}^{-1}$ (year 4-20) and $22.5 \text{ bale ha}^{-1}$ (year 3)
- JB6, wet and dry climate: $69.1 \text{ bale ha}^{-1}$ (year 4-20) and $24.9 \text{ bale ha}^{-1}$ (year 3)

Apart from the adjustment for diesel consumption and the conversion of flows from bale based to ha based, another modification was performed to adapt the process to the present study. This consisted to take out the polyethylene wrapping foil and plastic extrusion inputs.

Harvesting of willow occurs during vegetative rest periods, i.e. in the period around November to February. There are different harvesting techniques (Nordh and Dimitriou, 2003) and these are constantly improving, as this is a relatively new energy crop. As for *Miscanthus*, the Ecoinvent process “Chopping, maize/CH U”, is used as a best proxy for modelling the impacts related to harvesting (this basically is to reflect the tractor use and machinery use). The diesel consumption is estimated at 70 L ha^{-1} , based on a JF Z20 Hydro/E harvester from NYVRAA Bioenergy (NYVRAA Bioenergy, 2011). Therefore, a consumption of 63.0 L ha^{-1} (soil JB3) and 70.0 L ha^{-1} (soil JB6) is used.

When straw is not incorporated, the harvested field needs to be swath and then the cut straw can be bale. The process used for swathing is the Ecoinvent process “swath, by rotary windrower/CH U”, which assumes a diesel

consumption of 2.94 kg ha⁻¹. This is the value considered for soil JB6, and is adjusted by a factor 0.9 for soil JB3, meaning a diesel consumption of 2.65 kg ha⁻¹ for soil JB3. It is the whole ha that needs to be swath.

For baling the straw, the process “Baling/ CH U” from the Ecoinvent database has been used. It needs to be adjusted just as described above for harvesting of spring *Miscanthus*, i.e. by determining the number of bales per ha of the main crop. As a result, all values in the Ecoinvent process will need to be multiplied by:

- Spring barley straw, JB3: 3.956 bale ha⁻¹ (with or without catch crop)
- Spring barley straw, JB6: 4.508 bale ha⁻¹
- Winter wheat straw, JB3: 5.221 bale ha⁻¹
- Winter wheat straw, JB6: 6.325 bale ha⁻¹.

The values above were calculated based on the straw yield and the ratio FW:DM. A factor of 0.23 has also been applied, as recommended by Nemecek & Kägi (2007), since it is straw bales that it is loaded and not silage bale, for what the process was originally design for.

(Example for spring barley, JB3: 2.34 t DM straw/ha *bale/160 kg FW * kg FW/0.85 kg DM * 1000 kg/t * 0.23 = 3.956 bale ha⁻¹)

The diesel consumption assumed is 0.743 kg bale⁻¹, and is not adjusted.

For loading, the Ecoinvent process “Loading bales” is used, which is also expressed per bale. The diesel consumption assumed is 0.0811 kg bale⁻¹. The transformation per ha is done as for above, but without the 0.23 factor, as recommended by Nemecek & Kägi (2007). This results to:

- Spring barley straw, JB3: 17.2 bale ha⁻¹ (with or without catch crop)
- Spring barley straw, JB6: 19.6 bale ha⁻¹
- Winter wheat straw, JB3: 22.7 bale ha⁻¹
- Winter wheat straw, JB6: 27.5 bale ha⁻¹.

The process used for ryegrass harvesting is the same as for straw, i.e. the grass is swath and baled, and bales are loaded afterwards. For baling, the number of bale per ha is:

- Ryegrass, on JB3: 346.5 bale ha⁻¹
- Ryegrass, on JB6: 306.25 bale ha⁻¹.

For loading bales, the same values, in terms of bale ha⁻¹, applies. (The factor 0.23 does not apply here since it is the equivalent of silage).

The harvesting of beet tops is considered as for common silage, i.e. bales are made from the tops (process baling) and these are afterwards loaded. For baling, the number of bale per ha is:

- Sugar beet top, on JB3 and JB6: 84.4 bale ha⁻¹.

For loading bales, the same values, in terms of bale ha⁻¹, applies. (The factor 0.23 does not apply here since it is the equivalent of silage).

10. Transport from field to farm

The transport distance considered between the farm (i.e. where the machinery, slurry and other inputs are stored, as well as the crops products) and the field is estimated as 1 km, based on the Ecoinvent database. Loaded inputs, like slurry, mineral fertilisers or pesticides that needs to be transported to the field are accounted for through their respective processes (e.g. slurry spreading process, fertilizing process and application of pesticides), assuming this 1 km distance.

The process “transport, tractor and trailer” from the Ecoinvent database is used for every operation where a load needs to be transported between the field and the farm. This applies for the harvested grain, the harvested straw as well as the harvested cuttings and rhizomes. The Ecoinvent process “transport, tractor and trailer” takes into account that the loaded tractor is loaded at full capacity only in one direction and comes back empty. The process is expressed in tkm, i.e. one tonne of good transported over 1 km. Table S13 presents the tonnes of fresh product to be transported for each product to transport. A diesel consumption of 0.0436 kg per tkm is considered.

Table S13. Tonnes of fresh product to be transported, for each product to be transported

Product to transport	Tonnes of fresh product $\text{ha}^{-1} \text{y}^{-1}$ (to transport over 1 km)	Comment
Spring barley, JB3	5	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Spring barley, JB6	5.7	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Winter wheat, JB3	6.6	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Winter wheat, JB6	8.0	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Sugar beet	56.6	Considering 0.22 t dry weight t^{-1} fresh weight [*]
Silage maize, JB3	38.87	Considering 0.31 t dry weight t^{-1} fresh weight [*]
Silage maize, JB6	38.5	Considering 0.31 t dry weight t^{-1} fresh weight [*]
Ryegrass, JB3	55.42	Considering 0.18 t dry weight t^{-1} fresh weight [*]
Ryegrass, JB6	48.98	Considering 0.18 t dry weight t^{-1} fresh weight [*]
Willow, JB3, wet	21.2	Assuming 0.50 t dry weight t^{-1} fresh weight [†]
Willow, JB3, dry	14.2	Assuming 0.50 t dry weight t^{-1} fresh weight [†]
Willow, JB6, wet	25.44	Assuming 0.50 t dry weight t^{-1} fresh weight [†]
Willow, JB6, dry	21.62	Assuming 0.50 t dry weight t^{-1} fresh weight [†]
<i>Miscanthus</i> , autumn, wet, 4-20	34.66	Assuming 0.44 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , autumn, dry, JB3, 4-20	29.45	Assuming 0.44 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , autumn, dry, JB6, 4-20	34.66	Assuming 0.44 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , spring, wet, JB3, 4-20	11.76	Assuming 0.85 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , spring, dry, JB3, 4-20	10.00	Assuming 0.85 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , spring, JB6, 4-20	11.06	Assuming 0.904 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , autumn, wet, y3	12.48	Only 60 % harvest. Assuming 0.44 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , autumn, dry, JB3, y3	10.61	Only 60 % harvest. Assuming 0.44 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , autumn, dry, JB6, y3	12.48	Only 60 % harvest. Assuming 0.44 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , spring, wet, JB3, y3	4.23	Only 60 % harvest. Assuming 0.85 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , spring, wet, JB6, y3	3.98	Only 60 % harvest. Assuming 0.904 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , spring, dry, JB3, y3	3.60	Only 60 % harvest. Assuming 0.85 t dry weight t^{-1} fresh weight [‡]
<i>Miscanthus</i> , spring, dry, JB6, y3	3.98	Only 60 % harvest. Assuming 0.904 t dry weight t^{-1} fresh weight [‡]
Spring barley straw, JB3	2.75	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Spring barley straw, JB6	3.14	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Winter wheat straw, JB3	3.64	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Winter wheat straw, JB6	4.4	Considering 0.85 t dry weight t^{-1} fresh weight [*]
Sugar beet top	13.5	Considering 0.12 t dry weight t^{-1} fresh weight [*]
Rhizomes	3	This is for a mixture of rhizomes and stones. Value based on a personal communication with Jens B. Kjeldsen, Aarhus University (2010).
Cuttings	1.5	Based on a planting density of 15000 cuttings ha^{-1} , and considering that each cutting has a weight of 0.1 kg.

^{*}Based on Møller *et al.* (2002).

[†]Based on Heller *et al.* (2003); Jensen *et al.* (2009); Ledin (1996) and Mleczek *et al.* (2010).

[‡]Based on Kristensen (2003).

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Appendix 3. Primary yields data details for willow and *Miscanthus*

1. Willow primary yield

For willow, yield was adapted from Lærke *et al.* (2010):

- In Lærke *et al.* (2010), an average yield of 12.5 ton DM per hectare can be estimated from all data presented in figure 1, for the hybrid “Bjørn”. Yield for willow cultivated on soil JB3, under wet climate was estimated to be 85 % of this average yield. This assumes that commercial yield would be reduced of 15 % when compared to research yield, due to unproductive turning areas at field margins as well as to harvest losses.
- Yield for willow cultivated on soil JB3, under dry climate, is estimated to be 67 % of the yield under wet climate, for the same soil type. This is based on Mortensen *et al.* (1998) as well as on unpublished data showing the sensitivity of willow yield to different drought conditions.
- Yield for willow cultivated on soil JB6, under wet climate, is assumed to be the same as the yield on JB3 (wet climate) plus an increase corresponding to what would the increase of yield would be if irrigation is performed (on a JB3 soil, under wet climate). Based on unpublished data from the Faculty of Agricultural Sciences, Aarhus University (Denmark), such irrigation would increase the yield by 20 %. Therefore, the yield assumed for willow on soil JB6 (wet climate), corresponds to the yield on soil JB3 (wet climate) plus 20 %.
- Yield for willow cultivated on soil JB6, under dry climate, is assumed to be 85 % of the yield obtained under dry climate.

2. *Miscanthus* primary yield

Yield data for *Miscanthus* (spring and autumn harvest) are adapted from Olesen *et al.* (2001):

- For year 4 to 20 (established *Miscanthus*):
 - Soil JB3, wet climate: the yield is estimated as the average of the values for soils JB1 and JB4 presented in Olesen *et al.* (2001) (values for year 4-20). This corresponds to 15.25 t DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and to 10 t DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.
 - Soil JB3, dry climate: the yield is estimated as 85 % of the above calculated value. This assumes that there is a 15 % yield decrease for *Miscanthus* under the dry climate. This is lower than for willow because *Miscanthus* uses less water than willow. This gives a yield of 12.96 t DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and of 8.5 t DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.

- Soil JB6, for both climates: the yield is set identical to the yield on soil JB3, wet climate (15.25 t DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and to 10 t DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*). For wet climate, there is no increase as compared to what occurs on soil JB3 as the effect of “extra water availability” on JB6 is assumed to be compensated by the fact that *Miscanthus* benefits from a warmer soil on JB3. For dry climate, the yield is also set the same as for JB3, wet climate as *Miscanthus* probably seldom lacks water on a dry sandy loam soil.
- For year 1: No yield, as this is prior to plantation.
- Year 2 and 3: same procedure as for year 4-20. The values obtained with this procedure correspond to the total primary yield. However, for year 2, this yield is not harvested, and for year 3, only 60 % of this yield is harvested. This corresponds to:
 - Soil JB3, wet, soil JB6, wet and soil JB6, dry, year 2: 5.5 t DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 4.0 t DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus* (total yield that is not harvested)
 - Soil JB3, wet, soil JB6, wet and soil JB6, dry, year 3: 9.2 t DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 6.0 t DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus* (total yield, 60 % of this is harvested)
 - Soil JB3, dry, year 2: 4.7 t DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 3.4 t DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus* (total yield that is not harvested)
 - Soil JB3, dry, year 3: 7.8 t DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 5.1 t DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus* (total yield, 60 % of this is harvested)

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Appendix 4. Estimates for above and below ground residues of perennial crops

1. Ryegrass

The above- and below-ground residues input for perennial ryegrass has been estimated to 4.15 Mg DM ha⁻¹ y⁻¹ and 8.3 Mg DM ha⁻¹ y⁻¹, respectively. This is based on an overall C input to soil (i.e. above- + below-ground residues) estimated to 5.6 t C ha⁻¹ y⁻¹ (Petersen, 2010), and on the assumption that below-ground and above-ground residues are distributed according to a ratio 2:1 (IPCC, 2006). Assuming a content of 0.45 kg C kg⁻¹ DM (see main paper, section *C and N returned to soil from non-harvested residues*), the total of above- and below-ground C input to soil is 12.44 Mg DM ha⁻¹ y⁻¹. Since below-ground residues here correspond to above-ground residues multiplied by 2, the above-ground residues can be calculated as 12.44 Mg DM ha⁻¹ y⁻¹ divided by 3, which amounts to 4.15 Mg DM ha⁻¹ y⁻¹ of above-ground residues. The below-ground residues are then the double of this, i.e. 8.30 Mg DM ha⁻¹ y⁻¹.

2. Miscanthus

For *Miscanthus*, data for residual above ground biomass are, as for primary yield, mostly based on Olesen *et al.* (2001), as these values are based on Danish experiments. Accordingly:

- Year 1: the above ground biomass for year 1 is 0 as this is before the plantation.
- Year 2:
 - For JB3, wet climate: the average of the values presented by Olesen *et al.* (2001) for soil JB1 and JB4 for year 2 is considered, i.e. 2.04 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 3.54 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.
 - For JB3, dry climate: the above ground residual biomass yield was assumed to decrease of 15 % compared the value calculated for JB3, wet climate, as this assumption was made when establishing the primary yield. This gives an above ground residual biomass of 1.73 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 3.01 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.
 - For JB6, both climates: same values as for JB3, wet climate assumed, conformingly to the assumptions made for primary yield.
- Year 3: Same procedure as for year 2. This gives:
 - For JB3, wet climate: 3.51 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 5.76 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.
 - For JB3, dry climate: 2.98 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 4.90 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.
 - For JB6, both climates: 3.51 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 5.76 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.
- Year 4 to 20: Same procedure as for year 2 and 3. This gives:
 - For JB3, wet climate: 5.63 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 10.88 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.

- For JB3, dry climate: 4.79 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 9.25 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.
- For JB6, both climates: 5.63 Mg DM ha⁻¹ y⁻¹ for autumn harvested *Miscanthus* and 10.88 Mg DM ha⁻¹ y⁻¹ for spring harvested *Miscanthus*.

The below ground biomass was estimated as 16 % of the total above ground biomass (residues plus harvest), conformingly with Olesen *et al.* (2001). For years with no or partial harvest, this takes into account the entire primary yield.

Table S14 summarizes all data about residual above and below ground biomass for *Miscanthus*.

Table S14. Summary of above and below ground residual biomass amounts for *Miscanthus*

<i>Miscanthus</i> type	Soil type	Climate type	Year in crop life cycle	Biomass amount (Mg DM ha ⁻¹ y ⁻¹)	
				Above ground	Below ground
Autumn harvest	JB3, JB6	wet, dry	1	0	0
Autumn harvest	JB3	wet	2	2.04	1.21
Autumn harvest	JB3	wet	3	3.51	2.03
Autumn harvest	JB3	wet	4-20	5.63	3.34
Autumn harvest	JB3	dry	2	1.73	1.03
Autumn harvest	JB3	dry	3	2.98	1.72
Autumn harvest	JB3	dry	4-20	4.79	2.84
Autumn harvest	JB6	wet, dry	2	2.04	1.21
Autumn harvest	JB6	wet, dry	3	3.51	2.03
Autumn harvest	JB6	wet, dry	4-20	5.63	3.34
Spring harvest	JB3, JB6	wet, dry	1	0	0
Spring harvest	JB3	wet	2	3.54	1.21
Spring harvest	JB3	wet	3	5.76	1.88
Spring harvest	JB3	wet	4-20	10.88	3.34
Spring harvest	JB3	dry	2	3.01	1.03
Spring harvest	JB3	dry	3	4.90	1.60
Spring harvest	JB3	dry	4-20	9.25	2.84
Spring harvest	JB6	wet, dry	2	3.54	1.21
Spring harvest	JB6	wet, dry	3	5.76	1.88
Spring harvest	JB6	wet, dry	4-20	10.88	3.34

3. Willow

For willow, the DM from non-harvested above ground biomass (NHAG) is estimated as the DM from leaves (*L*) plus the DM from woody material (*WM*) lost from the trees (e.g. branches and twigs). This is illustrated in equation (S1):

$$\mathbf{NHAG = L + WM} \quad \text{(Equation S1)}$$

For the woody biomass DM, it is anticipated that the loss during harvest corresponds to 7.5 % of the total above ground biomass production. This total above ground biomass production is estimated by dividing the primary yield by 92.5 %, i.e. assuming that the anticipated primary yield is only 92.5 % of the full potential yield, because of losses. This could have been divided by 0.85 instead, then accounting for the turning areas, but this is not done here because it is the ambition of the present database to be as disaggregated as possible. The DM from woody material is therefore estimated as in equation (S2):

$$\mathbf{WM = 0.075 \times \frac{PY}{0.925}} \quad \text{(Equation S2)}$$

Where PY is the primary yield (in terms of DM per ha).

The DM from leaf is estimated based on the model developed by Lindroth & Båth (1999). Based on Lindroth & Båth (1999), it is assumed that 20 % of the total biomass production (TBP) is going to leaves (f_L), and 25 % to roots (f_R). Lindroth & Båth (1999), in their equation 2, express the total biomass production as in equation (S3) below:

$$\mathbf{TBP = \frac{PY}{(1 - f_L - f_R)}} \quad \text{(Equation S3)}$$

This equation is used to estimate the amount of DM from the leaves, but it needs to be improved. In fact, this equation does not include the DM from woody material. To include the woody material biomass, and taking into account the value for f_L (0.2) and f_R (0.25), equation (S3) may be rewritten as:

$$\mathbf{TBP = \frac{PY + WM}{0.55}} \quad \text{(Equation S4)}$$

Based on the definition and value of f_L , equation (S1) may be rewritten as:

$$\mathbf{NHAG = 0.20TBP + WM} \quad \text{(Equation S5)}$$

Combining equations (S5) with equation (S2) and equation (S4) allows calculating the non-harvestable above ground biomass from the primary yield only:

$$\mathbf{NHAG = \left(0.075 \times \frac{PY}{0.925} \right) + \frac{0.20}{0.55} \left[PY + \left(0.075 \times \frac{PY}{0.925} \right) \right]} \quad \text{(Equation S6)}$$

The below ground biomass (BG) is calculated based on the definition and value of f_R (i.e. the fraction of total biomass production going to roots; 25 % based on Lindroth & Båth, 1999) as well as on equation (S4) and (S2). This is presented in equation (S7):

$$\mathbf{BG = \frac{0.25}{0.55} \left[PY + \left(0.075 \times \frac{PY}{0.85} \right) \right]} \quad \text{(Equation S7)}$$

Based on equations S6 and S7, the DM in non-harvestable above ground biomass as well as in below ground biomass for willow is presented in Table S15.

TableS15. DM in non harvestable above ground biomass and below ground biomass for willow, for soil JB3 and JB6, under wet and dry climates

Crop	Soil type	Climate	Non harvestable above ground biomass (Mg DM ha ⁻¹ y ⁻¹)	Below ground biomass (Mg DM ha ⁻¹ y ⁻¹)
Willow	JB3	Wet	5.03	5.21
Willow	JB3	Dry	3.37	3.49
Willow	JB6	Wet	6.03	6.25
Willow	JB6	Dry	5.13	5.31

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Appendix 5. N leaching estimates for *Miscanthus* and willow

For *Miscanthus*, values for N leaching are based on Olesen *et al.* (2001).

- JB3, wet climate, year 4-20, spring and autumn harvest: 14 kg NO₃-N ha⁻¹ y⁻¹ (value for soil JB1 in Olesen *et al.*, 2001)
- JB3, dry climate, year 4-20, spring and autumn harvest: an increase of 20 % of the leaching under wet climate is estimated, resulting in a leaching of 17 kg NO₃-N ha⁻¹ y⁻¹.
- JB6, wet and dry climate, year 4-20, spring and autumn harvest: 10 kg NO₃-N ha⁻¹ y⁻¹ (value for soil JB4 in Olesen *et al.*, 2001)
- JB3, wet climate, year 2 and year 3, spring and autumn harvest: 89 kg NO₃-N ha⁻¹ y⁻¹ for year 2 and 17 kg NO₃-N ha⁻¹ y⁻¹ for year 3 (respective values for year 1 and year 2 of Olesen *et al.* (2001) for JB1).
- JB3, dry climate, year 2 and year 3, spring and autumn harvest: same procedure as for year 4-20 (i.e. leaching of 106.8 kg NO₃-N ha⁻¹ y⁻¹ for year 2 and 20.4 kg NO₃-N ha⁻¹ y⁻¹ for year 3)
- JB6, wet and dry climate, year 2 and 3, spring and autumn harvest: 75 kg NO₃-N ha⁻¹ y⁻¹ for year 2 and 20 kg NO₃-N ha⁻¹ y⁻¹ for year 3 (respective values for year 1 and year 2 of Olesen *et al.* (2001) for soil JB1).

Nitrate leaching values for willow are estimated to be the same as for *Miscanthus* (year 4-20).

For both *Miscanthus* and willow, N leaching is the highest in year where the first fertilisation occurred (here corresponding to year 2 of the life cycle in the case of *Miscanthus*, but often referred to as “year 1”). For willow, this higher N leaching occur on year 3 of the life cycle, and the values used are those of *Miscanthus* for year 2 (i.e. 89 kg NO₃-N ha⁻¹ y⁻¹ and 106.8 kg NO₃-N ha⁻¹ y⁻¹ on wet and dry JB3 and 75 kg NO₃-N ha⁻¹ y⁻¹ on wet and dry JB6).

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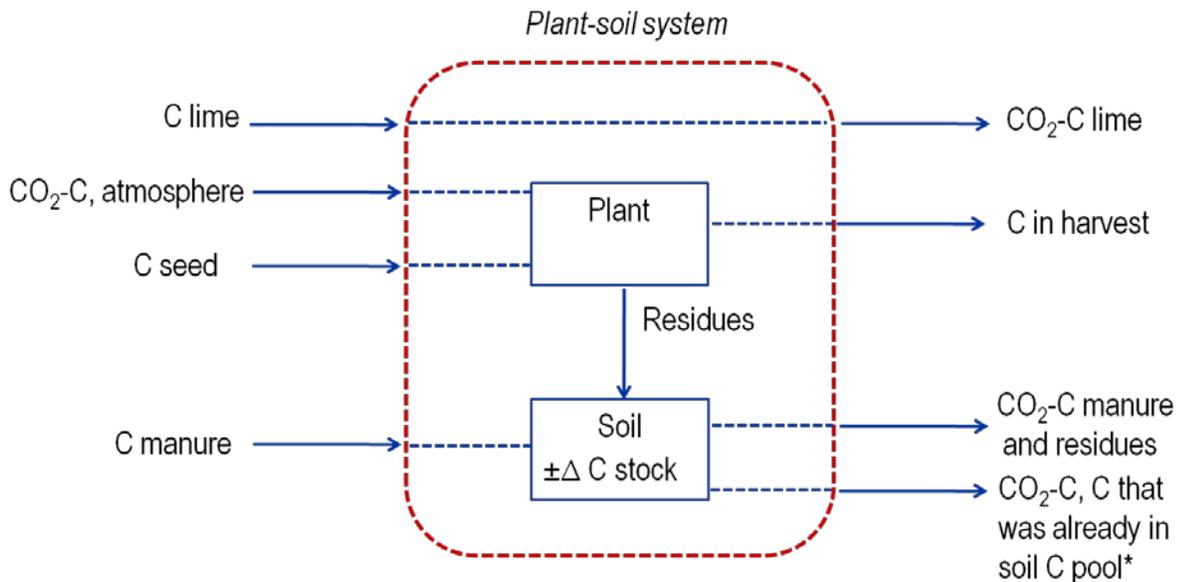
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Appendix 6. Carbon balance

1. Description of the system

The crop-soil systems considered involve 4 main C inputs: (i) lime; (ii) CO₂-C from the atmosphere; (iii) C from the seed/rhizome/cutting and (iv) C from the manure. Based on IPCC (2006), it is considered that all the C input from the applied lime ends up as CO₂-C emission to the atmosphere. The C from the seed/rhizome/cutting and the CO₂-C uptake from the atmosphere build up in the crop. Part of this C is harvested when the crop is harvested, and part had been lost through the non harvestable residues of the crop (above- and below-ground) which enter the soil C pool. Similarly, the C from the manure, which can of course not be sequestered in the plant, enters directly the soil C pool. Part of the C from these two C inputs will decay and end up as emissions of CO₂-C to the atmosphere, while the other part will contribute to increase the soil organic C stock. This applies for the systems where there is an increase of soil organic C. In some crop systems, the decay rate of C in the soil is superior than the C input from manure and residues. In these systems, not only all the C from manure and residues does decay to CO₂-C, but also some of the native C from the soil is decayed and emitted as CO₂-C to the atmosphere. These are the systems where there is a loss of soil C.

The soil-crop system is illustrated in Fig. S1.



* Facultative flow (when losses of native soil C only)

Figure S1. Illustration of the soil-crop system considered for the C balance

2. Calculation of the main flows

The flow of CO₂-C from the atmosphere was calculated as:

$$C_{\text{sequestered in plant}} = C_{\text{harvested}} + C_{\text{residues}} - C_{\text{seed}} \quad (\text{Equation S8})$$

The C from the seed was estimated assuming that 50 % of the seed/rhizome/cutting weight is C, and that seeds have a water content of 9 %, and rhizomes and cuttings of 40 %. For *Miscanthus* rhizomes, it was assumed that 40 % of the weight is stones. The data for the weight of rhizomes/cuttings are from Table S13, and the data for the amount of seed used from Table S3 (Appendix 2).

The C harvested is the C from both the primary yield and secondary yield, as found in Table 2. The C from residues is the C from both above- and below-ground residues, also found in Table 2.

The delta stock of soil C is obtained through the model C-TOOL, as described in the main paper. The results, for each crop and soil type combinations, are illustrated in Fig.2 of the main paper. When the delta stock of soil C is negative, it is considered that all this native C lost ends up as a CO₂-C emission to the atmosphere (CO₂-C from C that was already in the soil).

The CO₂-C from manure and residues was calculated as:

$$(i) \quad \text{CO}_2\text{-C}_{\text{manure and residues}} = C_{\text{manure}} + C_{\text{residues}} \quad (\text{when there is losses of soil C}) \quad (\text{Equation S9.1})$$

$$(ii) \quad \text{CO}_2\text{-C}_{\text{manure and residues}} = C_{\text{manure}} + C_{\text{residues}} - C_{\text{accumulated in soil}} \quad (\text{when there is gain of soil C}) \quad (\text{Equation S9.2})$$

3. C balances for all crop systems

Table S16 presents the C balance for all crop systems on sandy soil, for a wet climate, and Table S17 for sandy loam soil, also for a wet climate. Table S18 presents the results for willow and *Miscanthus* under a dry climate, for both soil types (only these crops are presented under a dry climate since they are the only crops for which the C balance differs for the different climates).

Table S16. Carbon balance for all crop systems on sandy soil, for a wet climate. All values in kg C ha⁻¹ y⁻¹. Eventual inconsistencies are due to rounding.

Crop systems	Spring barley (R)*	Spring barley (I)*	Spring barley & catch crop (R)*	Spring barley & catch crop (I)*	Winter wheat (R)*	Winter wheat (I)*	Willow (100 % slurry)	<i>Miscanthus</i> (autumn) Year 4-20	<i>Miscanthus</i> (spring) Year 4-20	Maize silage	Sugar beet (R)*	Sugar beet (I)*	Rye-grass	Willow (100 % mineral)
INPUTS														
C sequestered in plant	5087	5087	5716	5716	7402	7402	8926	10359	10359	7497	9094	9094	10082	8926
C seed	33	33	34	34	78	78	450	540	540	8	0.8	0.8	9	450
C manure	608	608	526	526	801	801	1158	434	289	782	593	593	1650	0
C lime	20	20	20	20	20	20	5	5	5	20	20	20	5	5
OUTPUTS														
CO ₂ -C, manure and residues	2764	3816	3312	4316	4303	5431	5240	4288	6080	2864	3358	4086	6595	4388
CO ₂ -C, lime	20	20	20	20	20	20	5	5	5	20	20	20	5	5
C primary yield	1913	1913	1913	1913	2525	2525	4770	6863	4500	5423	5603	5603	4491	4770
C secondary yield (when harvested)	1052	0	1052	0	1388	0	0	0	0	0	728	0	0	0
CO ₂ -C, from native soil C losses	248	51	149	0	0	0	0	0	0	218	138	0.5	0	0
DELTA SOIL C, ANNUALIZED OVER 20 YEARS														
Accumulation/losses of soil C	-248	-51	-149	48	65	326	524	183	609	-218	-138	-0.5	656	218

* R: straw removal; I: straw incorporation

Table S17. Carbon balance for all crop systems on sandy loam soil, for a wet climate. All values in kg C ha⁻¹ y⁻¹. Eventual inconsistencies are due to rounding.

Crop systems	Spring barley (R)*	Spring barley (I)*	Spring barley & catch crop (R)*	Spring barley & catch crop (I)*	Winter wheat (R)*	Winter wheat (I)*	Willow (100 % slurry)	<i>Miscanthus</i> (autumn) Year 4-20	<i>Miscanthus</i> (spring) Year 4-20	Maize silage	Sugar beet (R)*	Sugar beet (I)*	Rye- grass	Willow (100 % mineral)
INPUTS														
C sequestered in plant	5806	5806	6434	6434	8972	8972	10801	10359	10359	7423	9094	9094	9561	10801
C seed	37.6	37.6	38.9	38.9	94.3	94.3	450	540	540	7.7	0.8	0.8	8.3	450
C manure	550	550	468	468	777	777	1158	434	289	671	492	492	1568	0
C lime	20	20	20	20	20	20	5	5	5	20	20	20	5	5
OUTPUTS														
CO ₂ -C, manure and residues	3011	4211	3559	4759	5100	6461	6151	4471	6213	2733	3256	3985	6723	5316
CO ₂ -C, lime	20	20	20	20	20	20	5	5	5	20	20	20	5	5
C primary yield	2183	2183	2183	2183	3060	3060	5724	6863	4500	5369	5603	5603	3969	5724
C secondary yield (when harvested)	1200	0	1200	0	1683	0	0	0	0	0	728	0	0	0
CO ₂ -C, from native soil C losses	460	216	353	109	21	0	0	10	0	510	413	265	0	0
DELTA SOIL C, ANNUALIZED OVER 20 YEARS														
Accumulation/losses of soil C	-460	-216	-353	-109	-21	322	535	-10	476	-510	-413	-265	445	212

* R: straw removal; I: straw incorporation

Table S18. Carbon balance for *Miscanthus* and willow under dry climate, for sandy and sandy loam soils. All values in kg C ha⁻¹ y⁻¹. Eventual inconsistencies are due to rounding.

Soil type	SANDY SOIL (dry climate)				SANDY LOAM SOIL (dry climate)			
Crop systems	Willow (100 % slurry)	<i>Miscanthus</i> (autumn) Year 4-20	<i>Miscanthus</i> (spring) Year 4-20	Willow (100 % mineral)	Willow (100 % slurry)	<i>Miscanthus</i> (autumn) Year 4-20	<i>Miscanthus</i> (spring) Year 4-20	Willow (100 % mineral)
INPUTS								
C sequestered in plant	5832	8724	8724	5832	9113	10359	10359	9113
C seed	450	540	540	450	450	540	540	450
C manure	1158	434	289	0	1158	434	289	0
C lime	20	20	20	20	20	20	20	20
OUTPUTS								
CO ₂ -C, manure and residues	4030	3802	5308	3086	5505	4471	6261	4670
CO ₂ -C, lime	20	20	20	20	20	20	20	20
C primary yield	3196	5833	3825	3196	4865	6863	4500	4865
C secondary yield (when harvested)	0	0	0	0	0	0	0	0
CO ₂ -C, from native soil C losses	0	0	0	93	0	33	0	0
DELTA SOIL C, ANNUALIZED OVER 20 YEARS								
Accumulation/losses of soil C	214	64	421	-93	352	-33	428	28

* R: straw removal; I: straw incorporation

4. References

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Appendix 7. Calculation of NMVOC from photosynthesizing leaves

This category reflects the biogenic non-methane volatile organic compounds (NMVOC) emitted from photosynthesizing leaves of crops (particularly isoprene and monoterpene). The inclusion of NMVOC, though the data are rather uncertain, was judged important given the magnitude of the differences between the emissions of biogenic NMVOC from woody crops and arable crops. As an example, Jungbluth *et al.* (2007b) estimated, based on a model allowing to account for regional difference in Europe, an annual emission of 53.1 kg isoprene per ha for willow (willow-salix) while the annual emission for *Miscanthus* and wheat was of 21.6 and 20.1 kg isoprene per ha, respectively (in Switzerland) (increase of 155 % from the average of *Miscanthus* and wheat to the emission from willow). Annual monoterpene emissions estimated by Jungbluth *et al.* (2007b) amount to 2.7 kg monoterpene per ha for willow, 1.1 kg monoterpene per ha for *Miscanthus* and 1.0 kg monoterpene per ha for wheat. In the Ecoinvent database, biogenic emissions of NMVOC are not considered (Jungbluth *et al.* 2007).

Nielsen *et al.* (2009) used, in their estimation for the National greenhouse gas Inventory Report to the UNFCCC, an overall emission factor for biogenic NMVOC of 393 g NMVOC per ha for land with arable crops and 2120 g NMVOC per hectare for grassland.

The approach used in the German Inventory (Haenel *et al.* 2010) also ends up with an output in terms of NMVOC per ha per year, but is detailed in function of the crop type and the NMVOC type. This consists to multiply the primary yield (in DM ha⁻¹) by an emission factor for a given crop (kg NMVOC kg⁻¹ DM h⁻¹) by the fraction of year during which the crop is emitting. The crop emission factor and fraction of year data are given in table 11.7 of Haenel *et al.* (2010) and presented in Table S19. The calculation of NMVOC for this study is based on the methodology of Haenel *et al.* (2010), and results are presented in Table S20. *Miscanthus* and willow are not included in the data presented by Haenel *et al.* (2010).

Table S19. NMVOC emission factors presented by Haenel *et al.* (2010) and adapted in this study for estimating biogenic NMVOC. Empty cells are assumed to correspond to zero values.

Crop	Iso- prene (kg/kg DM*h)	Terpene (kg/kg DM*h)	Alcohols (kg/kg DM*h)	Alde- hydes (kg/kg DM*h)	Ketone (kg/kg DM*h)	Ethers and others (kg/kg DM*h)	Frac- tion of year emit- ting (y ⁻¹)	Primary yield (JB3) (this study) (Mg DM/ha)	Primary yield (JB6) (this study) (Mg DM/ha)	NMVOC (JB3) for this study (kg/ha)	NMVOC (JB6) for this study (kg/ha)
Wheat			8*10 ⁻¹⁰	2.8*10 ⁻⁹	2.2*10 ⁻⁹	5.1*10 ⁻⁹	0.3	5.61	6.8	0.161	0.195
Spring barley			8*10 ⁻¹⁰	2.8*10 ⁻⁹	2.2*10 ⁻⁹	5.1*10 ⁻⁹	0.3	4.25	4.85	0.122	0.139
Grass	2*10 ⁻¹⁰	6.3*10 ⁻⁹	7.5*10 ⁻¹⁰	1.3*10 ⁻⁹		1.8*10 ⁻⁹	0.5	9.98	8.82	0.452	0.400
Silage maize										0	0
Sugar beet										0	0

For 2008, Haenel *et al.* (2010) present an overall German emission factor of 0.13 kg NMVOC per ha (Table IEF 1001.11). The factors estimated above are a bit lower than the 0.393 kg NMVOC per hectare for arable crops presented by Nielsen *et al.* (2009), but for grass, it is far lower than the 2.120 kg/ha presented by Nielsen *et al.* (2009). In Switzerland, from the data of Jungbluth *et al.* (2007b), it appears that emissions are much higher. This could however be explained, at least partly, by the difference in terms of irradiation and sunshine hours, as well as primary yield.

For this study, the NMVOC are considered as a whole (not only for isoprene), and they are calculated based on the methodology presented by Haenel *et al.* (2010). This has the advantage to differentiate between crop types, crop yield, and consequently soil types. These data are judged to be representative for Danish conditions and are judged as best proxy under current data availability.

Based on the results of Jungbluth *et al.* (2007b), the emission factors for *Miscanthus* are assumed identical to those of wheat. For willow, emission factors are taken as those of grass, which give final NMVOC emissions close to the increase of 155 % as compared to emissions from wheat obtained by Jungbluth *et al.* (2007b). This approach is a rough estimate, but should be seen as an attempt to include NMVOC emissions from crops into LCA, which is seldom taken into account. The uncertainty for these emissions is estimated as a factor of 30 by Haenel *et al.* (2010), which is rather considerable.

Table S20 summarizes the NMVOC emissions for each crop types and soil types, and distinguishes for the weather in the case of willow and *Miscanthus* (based on primary yield data).

Table S20. NMVOC emissions for crops calculated in this study

Crop	NMVOC (soil JB3) (kg ha ⁻¹)	NMVOC (soil JB6) (kg ha ⁻¹)	Comments
Spring barley	0.122	0.139	From factors presented in Table S19.
Spring barley & catch crop	0.122	0.139	NMVOC from catch crop assumed negligible.
Winter wheat	0.161	0.195	From factors presented in Table S19.
<i>Miscanthus</i> (autumn harvest, year 4-20)	0.437 (wet) 0.371 (dry)	0.437 (wet & dry)	From factors presented in Table S19, for wheat.
<i>Miscanthus</i> (autumn harvest, year 3)	0.262 (wet) 0.223 (dry)	0.262 (wet & dry)	From factors presented in Table S19, for wheat.
<i>Miscanthus</i> (autumn harvest, year 2)	0.158 (wet) 0.134 (dry)	0.158 (wet & dry)	From factors presented in Table S19, for wheat.
<i>Miscanthus</i> (Spring harvest, year 4-20)	0.286 (wet) 0.243 (dry)	0.286 (wet & dry)	From factors presented in Table S19, for wheat.
<i>Miscanthus</i> (Spring harvest, year 3)	0.172 (wet) 0.146 (dry)	0.172 (wet & dry)	From factors presented in Table S19, for wheat.
<i>Miscanthus</i> (Spring harvest, year 2)	0.115 (wet) 0.097 (dry)	0.115 (wet & dry)	From factors presented in Table S19, for wheat.
Willow	0.481 (wet) 0.322 (dry)	0.576 (wet) 0.490 (dry)	From factors presented in Table S19, for grass.
Sugar beet	0	0	From factors presented in Table S19.
Silage maize	0	0	From factors presented in Table S19.
Permanent ryegrass	0.452	0.400	From factors presented in Table S19.

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Appendix 8. Calculation of phosphorus, copper and zinc losses

1. Phosphorus

Different approaches can be found in the literature as regarding how to treat the P losses to soil and water in LCA. Approaches used in recent studies are summarized in Table S21.

Table S21. Approaches inventoried from recent studies regarding the estimation of P losses in LCA

Reference	Assumption for P leaching	Remark
Nielsen & Wenzel (2007)	P leaching = 5 % of the net surplus application.	Rough estimation based on the Danish figures for P-surplus and P leaching to aquatic recipients (0.4 to 0.5 kg P ha ⁻¹ y ⁻¹), assuming that P surpluses are directly proportional to P leaching. Surplus application is estimated as: average P from manure application (30 kg P ha ⁻¹ y ⁻¹) minus average plant uptake (20 kg P ha ⁻¹ y ⁻¹).
Dalgaard (2007)	P leaching as phosphate = 2.9 % of P surplus.	Rough estimation based on the Danish figures for P-surplus and P emissions to aquatic recipients, assuming P surpluses are directly proportional to P leaching. Seen as an "average" for Danish agriculture (including both livestock and crop farms).
SenterNovem (2010)	P leaching as phosphate = 9 % of P surplus	Estimation based on a Dutch study from 1996.
Thomassen <i>et al.</i> (2009)	P leaching = 100% of P surplus	Assumption based on the Dutch context, based on the fact that most of the Dutch soils are phosphorus-saturated.
Hauschild & Potting (2005)	P leaching to soil = 0.10 kg per kg of P applied. (Annex 6.3) P leaching to water (inland and marine): 0.06*kg P leaching to soil (table 6.2).	This is in the case uptake and binding is unknown. If the actual amount of P leaving the top soil is known, it is recommended to use such figures.
Basset-Mens <i>et al.</i> (2007)	P losses to water = 1 % of P in runoff losses Runoff losses = 50 % of P excreted	Applies when P is from animal manure. Based on a reference from 1998.
Nemecek & Kägi (2007)	P leaching to ground water (kg/ha*y) = 0.07*[1+(0.2/80)*P ₂ O ₅ in slurry] P run-off to surface waters (kg/ha*y) = 0.175*[1+(0.2/80)*P ₂ O ₅ in mineral fertilisers + (0.7/80)* P ₂ O ₅ in slurry]	The factor 0.07 is for arable land. A factor of 0.06 is used for permanent pastures. The factor 0.175 is for arable land. A factor of 0.25 is used for (intensive) permanent pastures. The original formulation also includes a term for solid manure applied, but this is not considered in this project. A methodology for estimating P losses by water erosion is also presented.

From Table S21, it can be seen that most of the approaches for estimating P losses in recent studies are based on the amount of surpluses, implicitly assuming that the P losses are directly proportional to P surpluses. The validity of this assumption of linearity is questionable, and part of the P applied in non-surplus situations may also leak, depending on the P-status for the soil. The approach used in this study, as described in the main manuscript, consists to estimate the P losses as 5 % of the net surplus application for annual crops, and 2.5 % of the net surplus for perennial crops.

Table S22 shows the calculation of the P losses on soil JB3, and Table S23 on soil JB6, for all crops.

Table S22. Estimation of P losses from agricultural soils for soil JB3, for both residues management practices and for both climate types, unless otherwise specified.

P (kg ha ⁻¹ y ⁻¹)	Spring barley	Spring barley & Catch crop	Winter wheat	Willow* (100% slurry)	Misc. ^{††} , year 4 to 20 (autumn)	Misc. ^{††} , year 4 to 20 (spring)	Maize silage	Sugar beet	Ryegrass	Misc. ^{††} , Year 2 (both types)	Misc. ^{††} , Year 3 (both types)	Willow* (100% mineral)
P required, from law	22	22	22	15	15	15	44	43	36	15	15	22
Amount from slurry, total [A]	16.5	14.3	21.7	31.4	11.8	7.9	21.2	16.1	44.8	3.9	7.9	0
Amount from minerals [B]	5.5	7.7	0.3	0	3.2	7.1	22.8	26.9	0	11.1	7.1	22
Uptake, primary yield [‡] [C]	12.8	12.8 ^B	15.7	8.5 (w) 5.7 (d)	9.2 (w) 7.8 (d)	6.0 (w) 5.1 (d)	31.3	21.2	33.9	3.3 (w, autumn) 2.4 (w, spring) 2.8 (d, autumn) 2.0 (d, spring)	5.5 (w, autumn) 3.6 (w, spring) 4.7 (d, autumn) 3.1 (d, spring)	8.5 (w) 5.7 (d)
Uptake, secondary yield ⁰ [D]	2.1	2.1	2.8	0	0	0	0	4.4	0	0	0	0
Surplus, secondary yield harvested [E] = [(A+B)-C-D]	7.1	7.4	3.5	-	-	-	-	17.5	-	-	-	-
Surplus, no secondary yield or secondary yield left on-field [F] = [(A+B)-C]	9.3	9.5	6.3	22.9(w) 25.7 (d)	5.8 (w) 7.2 (d)	9.0 (w) 9.9 (d)	12.7	21.8	10.8	0.6 (w, autumn) 1.5 (w, spring) 1.1 (d, autumn) 1.9 (d, spring)	2.4 (w, autumn) 4.3 (w, spring) 3.2 (d, autumn) 4.8 (d, spring)	6.5 (w) 9.3 (d)
P losses to water recipients, secondary yield harvested [0.05*E]	0.36	0.37	0.18	-	-	-	-	0.87	-	-	-	-
P losses to water recipients, secondary yield left on-field or no secondary yield [0.05*F] or [0.025*F]	0.46	0.48	0.31	0.57(w) 0.64 (d)	0.15 (w) 0.18 (d)	0.23 (w) 0.25 (d)	0.63	1.09	0.27	0.02 (w, autumn) 0.04 (w, spring) 0.03 (d, autumn) 0.05 (d, spring)	0.06 (w, autumn) 0.11 (w, spring) 0.08 (d, autumn) 0.12 (d, spring)	0.16(w) 0.23 (d)

* w: wet; d: dry

† Misc. stems for *Miscanthus*

[‡]P in primary yield is taken from Møller *et al.* (2000): spring barley: 3 g P kg⁻¹ DM; winter wheat: 2.8 g P kg⁻¹ DM; silage maize: 2.6 g P kg⁻¹ DM; sugar beet: 1.7 g P kg⁻¹ DM; ryegrass: 3.4 g P kg⁻¹ DM. For *Miscanthus*, the value of 0.6 g P kg⁻¹ DM is used for both autumn and spring harvest, based on Beale and Long (1997). Content of P for willow seems to vary considerably with the site-specific P-availability, but 0.8 g P kg⁻¹ DM is a typical value for Danish soil (Sanders, 2010). To obtain the uptake in g P ha⁻¹ y⁻¹, these values are multiplied by the corresponding primary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Primary yields are taken from Table 2.

^βSince the catch crop is not harvested, there is no withdrawal of P considered from the catch crop.

^δP in secondary yield is taken from Møller *et al.* (2000): spring barley: 0.9 g P kg⁻¹ DM; winter wheat: 0.9 g P kg⁻¹ DM; sugar beet: 2.7 g P kg⁻¹ DM. To obtain the uptake in kg P ha⁻¹ y⁻¹, these values are multiplied by the corresponding secondary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Secondary yields are taken from Table 2.

Table S23. Estimation of P losses from agricultural soils for soil JB6, for both residues management practices and for both climate types, unless otherwise specified.

P (kg ha ⁻¹ y ⁻¹)	Spring barley	Spring barley & Catch crop	Winter wheat	Willow* (100% slurry)	Misc.** (year 4 to 20 (autumn))	Misc.** (year 4 to 20 (spring))	Maize silage	Sugar beet	Ryegrass	Misc.** (Year 2 (both types))	Misc.** (Year 3 (both types))	Willow* (100% mineral)
P required, from law	22	22	22	15	15	15	44	43	36	15	15	15
Amount from slurry, total [A]	14.9	12.7	21.1	31.4	11.8	7.9	18.2	13.3	42.5	3.9	7.9	0
Amount from minerals [B]	7.1	9.3	0.9	0	3.2	7.1	25.8	29.7	0	11.1	7.1	15
Uptake, primary yield [‡] [C]	14.6	14.6 ^β	19.0	10.2(w) 8.6 (d)	9.2 (w and d)	6.0 (w and d)	31.0	21.2	30.0	3.3 (w and d, autumn) 2.4 (w and d, spring)	5.5 (w and d, autumn) 3.6 (w and d, spring)	10.2(w) 8.6 (d)
Uptake, secondary yield ^δ [D]	2.40	2.40	3.37	0	0	0	0	4.37	0	0	0	0
Surplus, secondary yield harvested [E] = [(A+B)-C-D]	5.0	5.3	0	-	-	-	-	17.5	-	-	-	-
Surplus, no secondary yield or secondary yield left on-field [F] = [(A+B)-C]	7.5	7.7	3.0	21.2(w) 22.8(d)	5.8 (w and d)	9.0 (w and d)	13.0	21.8	12.5	0.6 (w and d, autumn) 1.5 (w and d, spring)	2.4 (w and d, autumn) 4.3 (w and d, spring)	4.8 (w) 6.4 (d)
P losses to water recipients, secondary yield harvested [0.05*E]	0.25	0.27	0	-	-	-	-	0.87	-	-	-	-
P losses to water recipients, secondary yield left on-field or no secondary yield [0.05*F] or [0.025*F]	0.37	0.39	0.15	0.53(w) 0.57 (d)	0.07 (w and d)	0.05 (w and d)	0.65	1.09	0.31	0.02 (w and d, autumn) 0.04 (w and d, spring)	0.06 (w and d, autumn) 0.11 (w and d, spring)	0.12 (w) 0.16 (d)

*w: wet; d: dry

[†]Misc. stems for *Miscanthus*.

[‡]P in primary yield is taken from Møller *et al.* (2000): spring barley: 3 g P kg⁻¹ DM; winter wheat: 2.8 g P kg⁻¹ DM; silage maize: 2.6 g P kg⁻¹ DM; sugar beet: 1.7 g P kg⁻¹ DM; ryegrass: 3.4 g P kg⁻¹ DM. For *Miscanthus*, the value of 0.6 g P kg⁻¹ DM is used for both autumn and spring harvest, based on Beale and Long (1997). Content of P for willow seems to vary considerably with the site-specific P-availability, but 0.8 g P kg⁻¹ DM is a typical value for Danish soil (Sanders, 2010). To obtain the uptake in g P ha⁻¹ y⁻¹, these values are multiplied by the corresponding primary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Primary yields are taken from Table 2.

^βSince the catch crop is not harvested, there is no withdrawal of P considered from the catch crop.

^δP in secondary yield is taken from Møller *et al.* (2000): spring barley: 0.9 g P kg⁻¹ DM; winter wheat: 0.9 g P kg⁻¹ DM; sugar beet: 2.7 g P kg⁻¹ DM. To obtain the uptake in kg P ha⁻¹ y⁻¹, these values are multiplied by the corresponding secondary yields (in Mg DM ha⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Secondary yields are taken from Table 2.

2. K losses

As acknowledged by Arienzo *et al.* (2009), the number of studies assessing the fate of K in the soil plant system following the application of organic fertilizer (from different sources) is rather limited. According to these authors, the risk of K losses from agricultural soils is rather low given the high propensity of K ions to be adsorbed by soil particles. Johnston & Goulding (2002), quoted in Arienzo *et al.* (2009), report that almost all exchangeable and non-exchangeable soil sites would have to be potassium-saturated before there is a serious risk of leaching. In two regional field studies carried out in the Netherlands, Griffioen (2001) measured an increase in the potassium adsorption ratio values (ratio K^+ to square root of sum of calcium, magnesium and iron (II); see Griffioen 2001 for details) in the ground waters below areas of agricultural land use as compared to pristine ground waters. These results suggest that losses of K from agricultural soil may be taking place. Askegaard & Eriksen (2008) explain that due to a low cationic exchange capacity on coarse sandy soils, there is a potentially high risk of K losses.

Askegaard & Eriksen (2008) measured, for spring barley crops fertilized by a KCl salt on a coarse sandy soil, losses of K varying between approximately 21 and 36 kg ha⁻¹ (values estimated from a graph). The authors calculated from their results that catch crops reduce the K losses by 28 %.

In another study, Askegaard *et al.* (2003) highlight the propensity of K losses for crops on sandy soils with less than 5 % of clay. The annual K losses measured by Askegaard *et al.* (2003) varied between 13 to 47 kg ha⁻¹ for crops (barley, grass clover and winter wheat) fertilized with manure on a coarse sandy soil (with less than 5 % clay). However, the authors highlight that a major part of K losses probably came from the straw residues. From their experimental results, Askegaard *et al.* (2003) significantly correlated the losses of K to the initial amount of exchangeable K in the soil.

Though the importance of understanding the fate of K on an agronomical perspective is recognized, K losses towards soils and waters is not a flow affecting any of the environmental impacts categories described in the Danish life cycle impact assessment method "EDIP" (Wenzel *et al.*, 1997; Stranddorf *et al.*, 2005; Potting & Hauschild, 2005; Hauschild & Potting, 2005). The fate of K (between soil and water) is therefore not considered further in this study.

3. Cu losses

It is considered that 100 % of the Cu surplus is lost. Part of this will bind with negatively charged particles of the soil, and another part may reach the aquatic recipients, depending, among others, on soil physicochemical properties and precipitations. Hao *et al.* (2008) highlight that heavy irrigation or precipitation combined with intensive organic fertilisation increase the propensity for heavy metal leaching.

Table S24 and S25 present the calculation of the Cu losses from agricultural soils on soil JB3 and JB6, respectively.

Table S24. Estimation of Cu losses from agricultural soils for soil JB3, for both residues management practices and for both climate types, unless otherwise specified.

Cu (kg ha ⁻¹ y ⁻¹)	Spring barley	Spring barley & Catch crop	Winter wheat	Willow ^{††} (100% slurry)	Misc. ^{‡‡} , year 4 to 20 (autumn)	Misc. ^{‡‡} , year 4 to 20 (spring)	Maize silage	Sugar beet	Ryegrass	Misc. ^{‡‡} , Year 2 (both types)	Misc. ^{‡‡} , Year 3 (both types)
Amount from slurry, total [A]	0.334	0.289	0.440	0.636	0.238	0.159	0.429	0.326	0.906	0.079	0.159
Uptake, primary yield ^β [B]	0.013	0.013 ⁰	0.011	0.064(w) 0.043(d)	0.034(w) 0.029(d)	0.022(w) 0.019(d)	0.060	0.050	0.070	0.012(autumn, w) 0.009(spring, w) 0.010(autumn, d) 0.007(spring, d)	0.020(autumn, w) 0.013(spring, w) 0.017(autumn, d) 0.011(spring, d)
Uptake, secondary yield ^α [C]	0.007	0.007	0.009	0	0	0	0	0.021	0	0	0
Surplus, secondary yield harvested [D] = [A-B-C]	0.314	0.269	0.419	-	-	-	-	0.255	-	-	-
Surplus, no secondary yield or secondary yield left on- field [E] = [A- B]	0.321	0.276	0.428	0.572(w) 0.593(d)	0.205(w) 0.210(d)	0.137(w) 0.140(d)	0.369	0.276	0.836	0.067(autumn, w) 0.071(spring, w) 0.069(autumn, d) 0.072(spring, d)	0.139(autumn, w) 0.146(spring, w) 0.142(autumn, d) 0.148(spring, d)
Cu losses to water recipients, secondary yield harvested [D*1]	0.314	0.269	0.419	-	-	-	-	0.255	-	-	-
Cu losses to water recipients, secondary yield left on- field or no secondary yield [E*1]	0.321	0.276	0.428	0.572(w) 0.593(d)	0.205(w) 0.210(d)	0.137(w) 0.140(d)	0.369	0.276	0.836	0.067(autumn, w) 0.071(spring, w) 0.069(autumn, d) 0.072(spring, d)	0.139(autumn, w) 0.146(spring, w) 0.142(autumn, d) 0.148(spring, d)

^{*}w: wet; d: dry

[†] willow fertilized with 100 % mineral does not have Cu surpluses since no Cu input is considered, and therefore no losses of Cu.

[‡] Misc. stems for *Miscanthus*.

^βCu in primary yield is taken from Møller *et al.* (2000): spring barley: 3 mg Cu kg⁻¹ DM; winter wheat: 2 mg Cu kg⁻¹ DM; silage maize: 5 mg Cu kg⁻¹ DM; sugar beet: 4 mg Cu kg⁻¹ DM; ryegrass: 7 mg Cu kg⁻¹ DM. For willow, values are taken from Mleczek *et al.* (2010), i.e. an average of 6.0559 mg Cu kg⁻¹ DM. This value is based on field trials, and corresponds to the

average of 11 different values (table 4 of Mleczek *et al.*, 2010). For *Miscanthus*, a value of 0.0022 g Cu kg⁻¹ DM is used, based on Smith & Slater (2010). To obtain the uptake in kg Cu ha⁻¹ y⁻¹, these values are multiplied by the corresponding primary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Primary yields are taken from Table 2.

^oSince the catch crop is not harvested, there is no withdrawal of Cu considered from the catch crop.

^λCu in secondary yield are taken from Møller *et al.* (2000): spring barley: 3 mg Cu kg⁻¹ DM; winter wheat: 3 mg Cu kg⁻¹ DM; sugar beet: 13 mg Cu kg⁻¹ DM. To obtain the uptake in kg Cu ha⁻¹ y⁻¹, these values are multiplied by the corresponding secondary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Secondary yields are taken from Table 2.

Table S25. Estimation of Cu losses from agricultural soils for soil JB6, for both residues management practices and for both climate types, unless otherwise specified.

Cu (kg ha ⁻¹ y ⁻¹)	Spring barley	Spring barley & Catch crop	Winter wheat	Willow ^{††} (100% slurry)	Misc. ^{‡‡} , year 4 to 20 (autumn)	Misc. ^{‡‡} , year 4 to 20 (spring)	Maize silage	Sugar beet	Ryegrass	Misc. ^{‡‡} , Year 2 (both types)	Misc. ^{‡‡} , Year 3 (both types)
Amount from slurry, total [A]	0.302	0.257	0.426	0.636	0.238	0.159	0.368	0.270	0.861	0.079	0.159
Uptake, primary yield ^β [B]	0.015	0.015 ^δ	0.014	0.077(w) 0.065(d)	0.034	0.022	0.060	0.050	0.06	0.012(autumn) 0.009 (spring)	0.020(autumn) 0.013 (spring)
Uptake, secondary yield ^λ [C]	0.008	0.008	0.011	0	0	0	0	0.021	0	0	0
Surplus, secondary yield harvested [D] = [A-B-C]	0.279	0.234	0.402	-	-	-	-	0.199	-	-	-
Surplus, no secondary yield or secondary yield left on- field [E] = [A- B]	0.287	0.242	0.413	0.559(w) 0.570(d)	0.205	0.137	0.309	0.220	0.799	0.067(autumn) 0.071 (spring)	0.139(autumn) 0.146 (spring)
Cu losses to water recipients, secondary yield harvested [D*1]	0.279	0.234	0.402	-	-	-	-	0.199	-	-	-
Cu losses to water recipients, secondary yield left on- field or no secondary yield [E*1]	0.287	0.242	0.413	0.559(w) 0.570(d)	0.205	0.137	0.309	0.220	0.799	0.067(autumn) 0.071 (spring)	0.139(autumn) 0.146 (spring)

* w: wet; d: dry

[†] Willow fertilized with 100 % mineral does not have Cu surpluses since no Cu input is considered, and therefore no losses of Cu.

[‡] Misc. stems for *Miscanthus*.

^β Cu in primary yield is taken from Møller *et al.* (2000): spring barley: 3 mg Cu kg⁻¹ DM; winter wheat: 2 mg Cu kg⁻¹ DM; silage maize: 5 mg Cu kg⁻¹ DM; sugar beet: 4 mg Cu kg⁻¹ DM; ryegrass: 7 mg Cu kg⁻¹ DM. For willow, values are taken from Mleczek *et al.* (2010), i.e. an average of 6.0559 mg Cu kg⁻¹ DM. This value is based on field trials, and corresponds to the average of 11 different values (table 4 of Mleczek *et al.*, 2010). For *Miscanthus*, a value of 0.0022 g Cu kg⁻¹ DM is used, based on Smith & Slater (2010). To obtain the uptake in kg Cu ha⁻¹ y⁻¹, these values are multiplied by the corresponding primary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Primary yields are taken from Table 2.

^δ Since the catch crop is not harvested, there is no withdrawal of Cu considered from the catch crop.

^λCu in secondary yield are taken from Møller *et al.* (2000): spring barley: 3 mg Cu kg⁻¹ DM; winter wheat: 3 mg Cu kg⁻¹ DM; sugar beet: 13 mg Cu kg⁻¹ DM. To obtain the uptake in kg Cu ha⁻¹ y⁻¹, these values are multiplied by the corresponding secondary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Secondary yields are taken from Table 2.

4. Zn losses

For estimating Zn losses, the same consideration as for Cu applies, i.e. it is considered that 100 % of the Zn surplus is lost.

Table S26 and S27 present the calculation of the Zn losses from agricultural soils on soil JB3 and JB6, respectively.

Table S26. Estimation of Zn losses from agricultural soils for soil JB3, for both residues management practices and for both climate types, unless otherwise specified.

Zn (kg ha ⁻¹ y ⁻¹)	Spring barley	Spring barley & Catch crop	Winter wheat	Willow ^{**} (100% slurry)	Misc. ^{**} , year 4 to 20 (autumn)	Misc. ^{**} , year 4 to 20 (spring)	Maize silage	Sugar beet	Ryegrass	Misc. ^{**} , Year 2 (both types)	Misc. ^{**} , Year 3 (both types)
Amount from slurry, total [A]	0.899	0.778	1.184	1.712	0.642	0.428	1.156	0.878	2.440	0.214	0.428
Uptake, primary yield ^b [B]	0.132	0.132 ²	0.146	0.583(w) 0.391(d)	0.459 (w) 0.390 (d)	0.301(w) 0.256(d)	0.856	0.896	0.399	0.166(autumn, wet) 0.120(spring, wet) 0.141(autumn, dry) 0.102(spring, dry)	0.275(autumn, wet) 0.181(spring, wet) 0.234(autumn, dry) 0.154(spring, dry)
Uptake, secondary yield ^a [C]	0.344	0.344	0.142	0	0	0	0	0.073	0	0	0
Surplus, secondary yield harvested [D] = [A-B- C]	0.423	0.302	0.896	-	-	-	-	0	-	-	-
Surplus, no secondary yield or secondary yield left on- field [E] = [A-B]	0.767	0.646	1.039	1.129(w) 1.322(d)	0.183(w) 0.252(d)	0.127(w) 0.172(d)	0.300	0	2.041	0.049(autumn, wet) 0.094(spring, wet) 0.073(autumn, dry) 0.112(spring, dry)	0.153(autumn, wet) 0.248(spring, wet) 0.194(autumn, dry) 0.275(spring, dry)
Zn losses to water recipients, secondary yield harvested [D*1]	0.423	0.302	0.896	-	-	-	-	0	-	-	-
Zn losses to water recipients, secondary yield left on- field or no secondary yield [E*1]	0.767	0.646	1.039	1.129(w) 1.322(d)	0.183(w) 0.252(d)	0.127(w) 0.172(d)	0.300	0	2.041	0.049(autumn, wet) 0.094(spring, wet) 0.073(autumn, dry) 0.112(spring, dry)	0.153(autumn, wet) 0.248(spring, wet) 0.194(autumn, dry) 0.275(spring, dry)

* w: wet; d: dry

[†] Willow fertilized with 100 % mineral does not have Zn surpluses since no Zn input is considered, and therefore no losses of Zn.

[‡] Misc. stems for *Miscanthus*.

^βZn in primary yield are taken from Møller *et al.* (2000): spring barley: 31 mg Zn kg⁻¹ DM; winter wheat: 26 mg Zn kg⁻¹ DM; silage maize: 71 mg Zn kg⁻¹ DM; sugar beet: 72 mg Zn kg⁻¹ DM; ryegrass: 40 mg Zn kg⁻¹ DM. For willow, values are taken from Mleczek *et al.* (2010), i.e. an average of 55.0464 mg Zn kg⁻¹ DM. This value is based on field trials, and corresponds to the average of 11 different values (table 4 of Mleczek *et al.*, 2010). For *Miscanthus*, a value of 0.0301 g Zn kg⁻¹ DM is used, based on Smith & Slater (2010). To obtain the uptake in kg Zn ha⁻¹ y⁻¹, these values are multiplied by the corresponding primary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Primary yields are taken from Table 2.

^δSince the catch crop is not harvested, there is no withdrawal of Zn considered from the catch crop.

^λZn in secondary yield are taken from Møller *et al.* (2000): spring barley: 147 mg Zn kg⁻¹ DM; winter wheat: 46 mg Zn kg⁻¹ DM; sugar beet: 45 mg Zn kg⁻¹ DM. To obtain the uptake in kg Zn ha⁻¹ y⁻¹, these values are multiplied by the corresponding secondary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Secondary yields are taken from Table 2.

Table S27. Estimation of Zn losses from agricultural soils for soil JB6, for both residues management practices and for both climate types, unless otherwise specified.

Zn (kg ha ⁻¹ y ⁻¹)	Spring barley	Spring barley & Catch crop	Winter wheat	Willow ^{**†} (100% slurry)	Misc. ^{**‡} , year 4 to 20 (autumn)	Misc. ^{**‡} , year 4 to 20 (spring)	Maize silage	Sugar beet	Ryegrass	Misc. ^{**‡} , Year 2 (both types)	Misc. ^{**‡} , Year 3 (both types)
Amount from slurry, total [A]	0.813	0.692	1.149	1.712	0.642	0.428	0.992	0.728	2.319	0.214	0.428
Uptake, primary yield ^β [B]	0.150	0.150 ^δ	0.177	0.700(w) 0.595(d)	0.459	0.301	0.847	0.896	0.35	0.166(autumn) 0.120(spring)	0.275(autumn) 0.181(spring)
Uptake, secondary yield ^α [C]	0.392	0.392	0.172	0	0	0	0	0.073	0	0	0
Surplus, secondary yield harvested [D] = [A-B- C]	0.271	0.149	0.800	-	-	-	-	0	-	-	-
Surplus, no secondary yield or secondary yield left on-field [E] = [A-B]	0.663	0.542	0.972	1.013(w) 1.118(d)	0.183	0.127	0.145	0	1.966	0.049(autumn) 0.094(spring)	0.153(autumn) 0.248(spring)
Zn losses to water recipients, secondary yield harvested [D*1]	0.271	0.149	0.800	-	-	-	-	0	-	-	-
Zn losses to water recipients, secondary yield left on-field or no secondary yield [E*1]	0.663	0.542	0.972	1.013(w) 1.118(d)	0.183	0.127	0.145	0	1.966	0.049(autumn) 0.094(spring)	0.153(autumn) 0.248(spring)

* w: wet; d: dry

† Willow fertilized with 100 % mineral does not have Zn surpluses since no Zn input is considered, and therefore no losses of Zn.

‡ Misc. stems for *Miscanthus*.

^βZn in primary yield are taken from Møller *et al.* (2000): spring barley: 31 mg Zn kg⁻¹ DM; winter wheat: 26 mg Zn kg⁻¹ DM; silage maize: 71 mg Zn kg⁻¹ DM; sugar beet: 72 mg Zn kg⁻¹ DM; ryegrass: 40 mg Zn kg⁻¹ DM. For willow, values are taken from Mleczek *et al.* (2010), i.e. an average of 55.0464 mg Zn kg⁻¹ DM. This value is based on field trials, and corresponds to the average of 11 different values (table 4 of Mleczek *et al.*, 2010). For *Miscanthus*, a value of 0.0301 g Zn kg⁻¹ DM is used, based on Smith & Slater (2010). To obtain the uptake in kg Zn ha⁻¹ y⁻¹, these values are multiplied by the corresponding primary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Primary yields are taken from Table 2.

^oSince the catch crop is not harvested, there is no withdrawal of Zn considered from the catch crop.

^λZn in secondary yield are taken from Møller *et al.* (2000): spring barley: 147 mg Zn kg⁻¹ DM; winter wheat: 46 mg Zn kg⁻¹ DM; sugar beet: 45 mg Zn kg⁻¹ DM. To obtain the uptake in kg Zn ha⁻¹ y⁻¹, these values are multiplied by the corresponding secondary yields (in Mg DM ha⁻¹ y⁻¹) and a unit conversion factor (1000 kg Mg⁻¹). Secondary yields are taken from Table 2.

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Appendix B. Supporting Information for Paper II

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.

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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 previously subtracted).

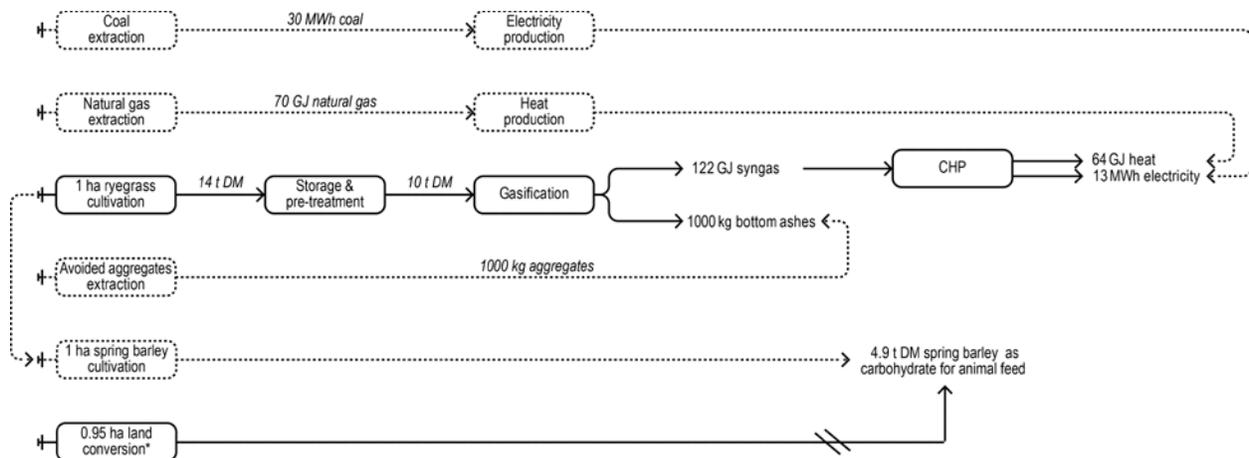


Figure S1. Process flow diagram for gasification of ryegrass. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Fly ashes are considered used as back-filling material in old salt mines, and the environmental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

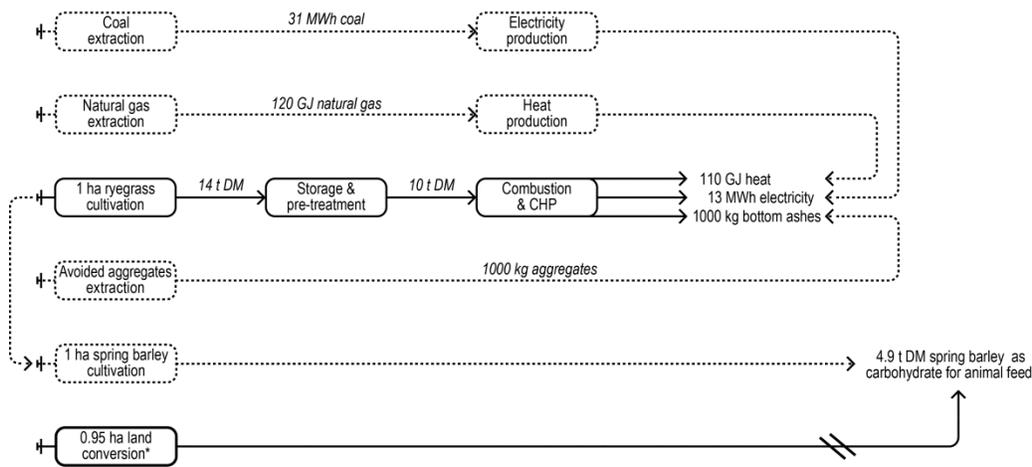


Figure S2. Process flow diagram for combustion of ryegrass. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Fly ashes are considered used as back-filling material in old salt mines, and the environmental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

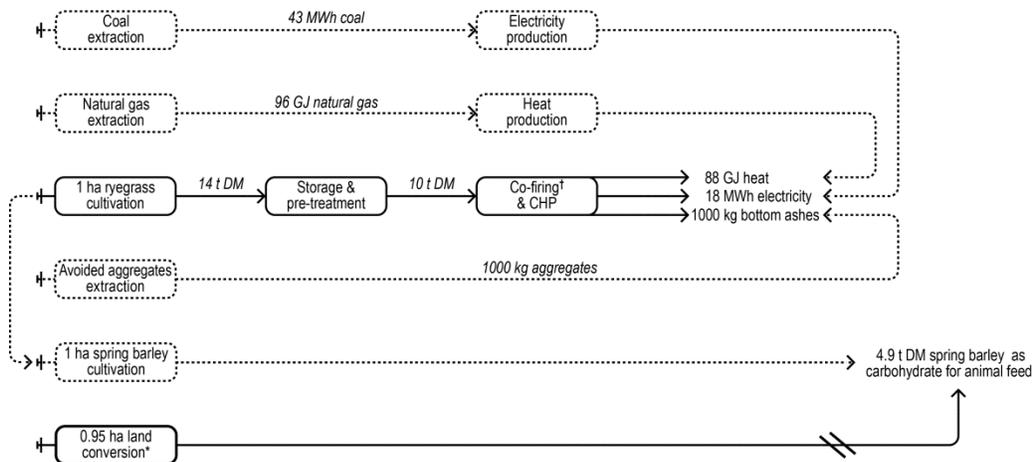


Figure S3. Process flow diagram for co-firing of ryegrass. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Based on the data from co-firing Danish plants, the coal that is used here would have otherwise 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.

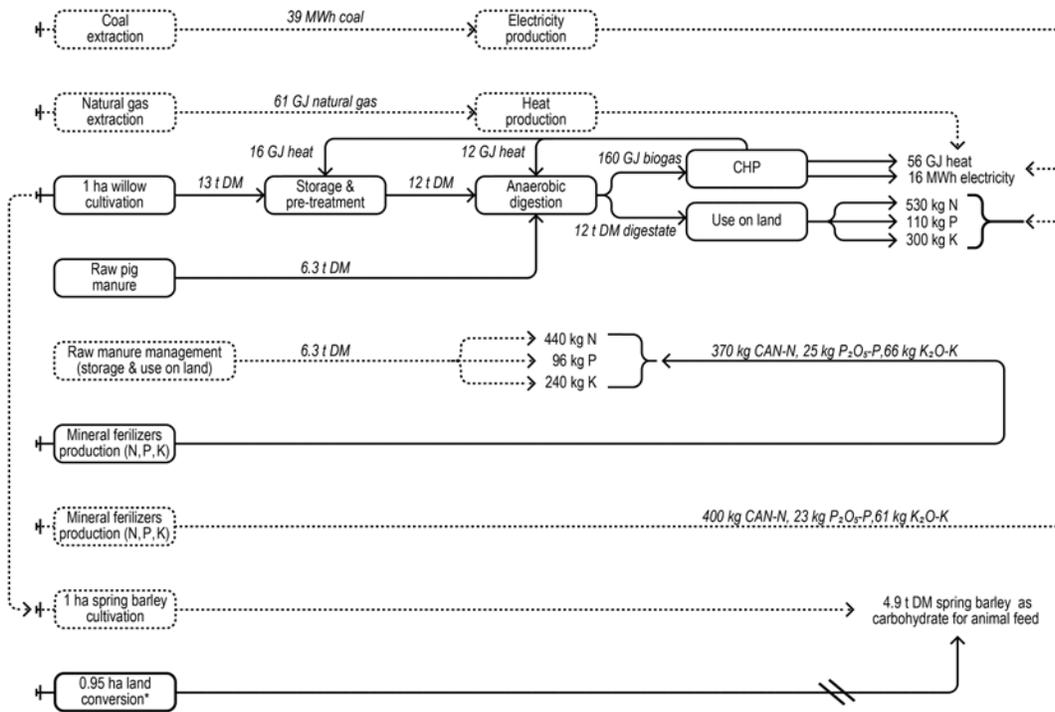


Figure S4. Process flow diagram for anaerobic co-digestion of willow with raw pig manure. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Values are rounded to 2 significant digits.

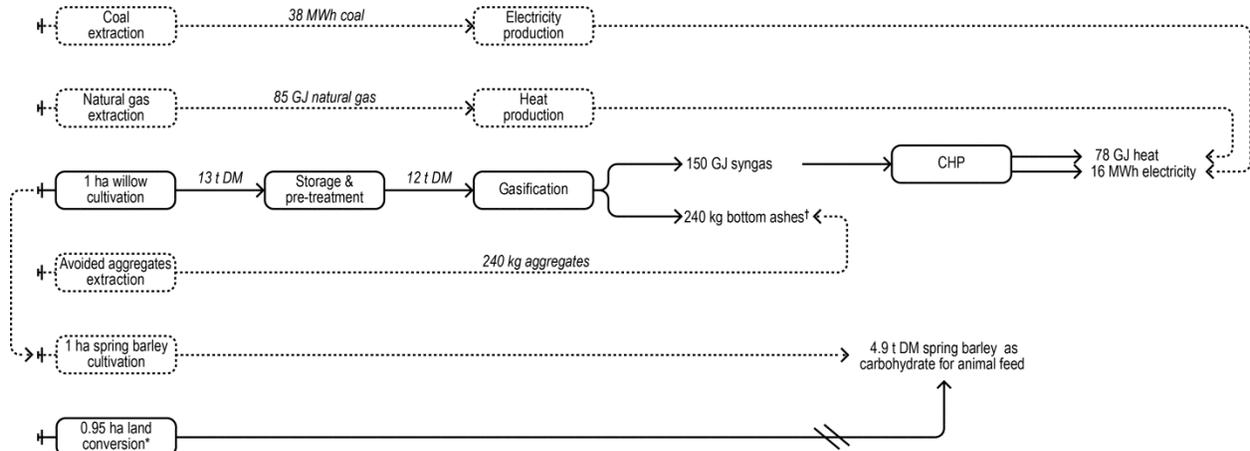


Figure S5. Process flow diagram for gasification of willow. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Fly ashes are considered used as back-filling material in old salt mines, and the environmental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

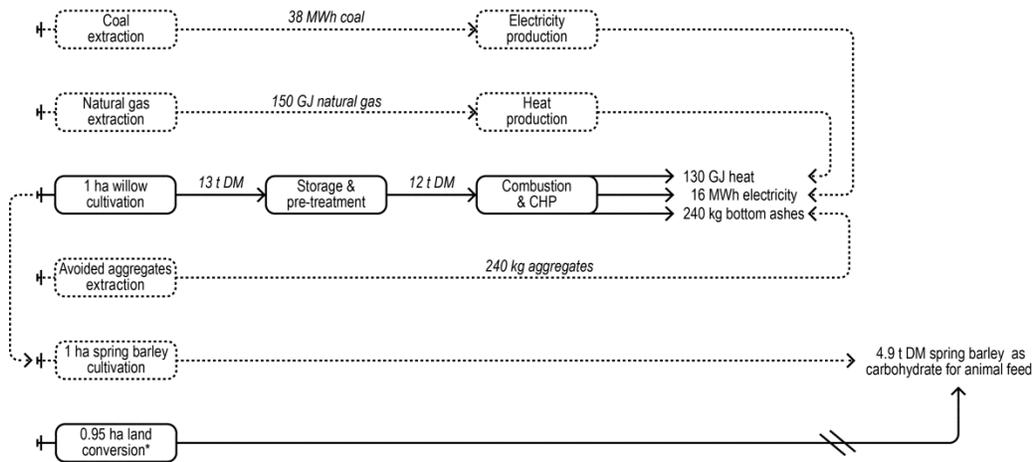


Figure S6. Process flow diagram for combustion of willow. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Fly ashes are considered used as back-filling material in old salt mines, and the environmental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

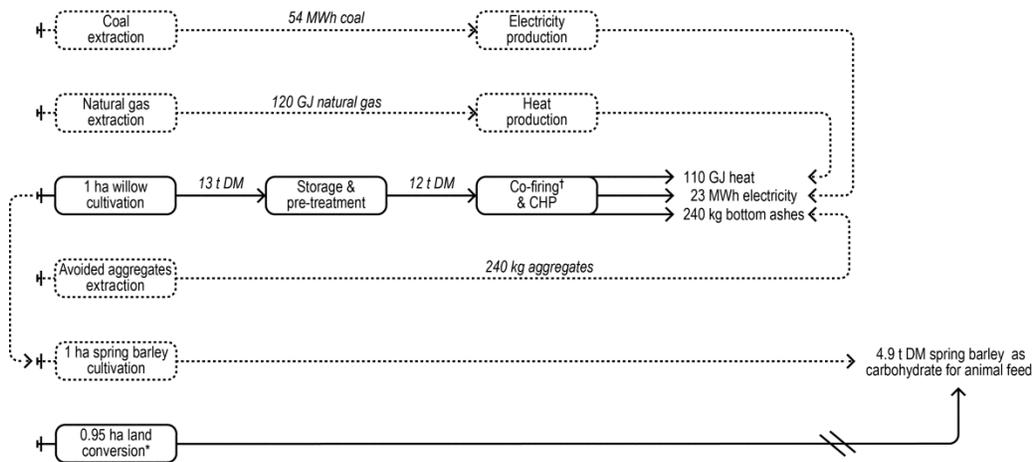


Figure S7. Process flow diagram for co-firing of willow. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Based on the data from co-firing Danish plants, the coal that is used here would have otherwise 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.

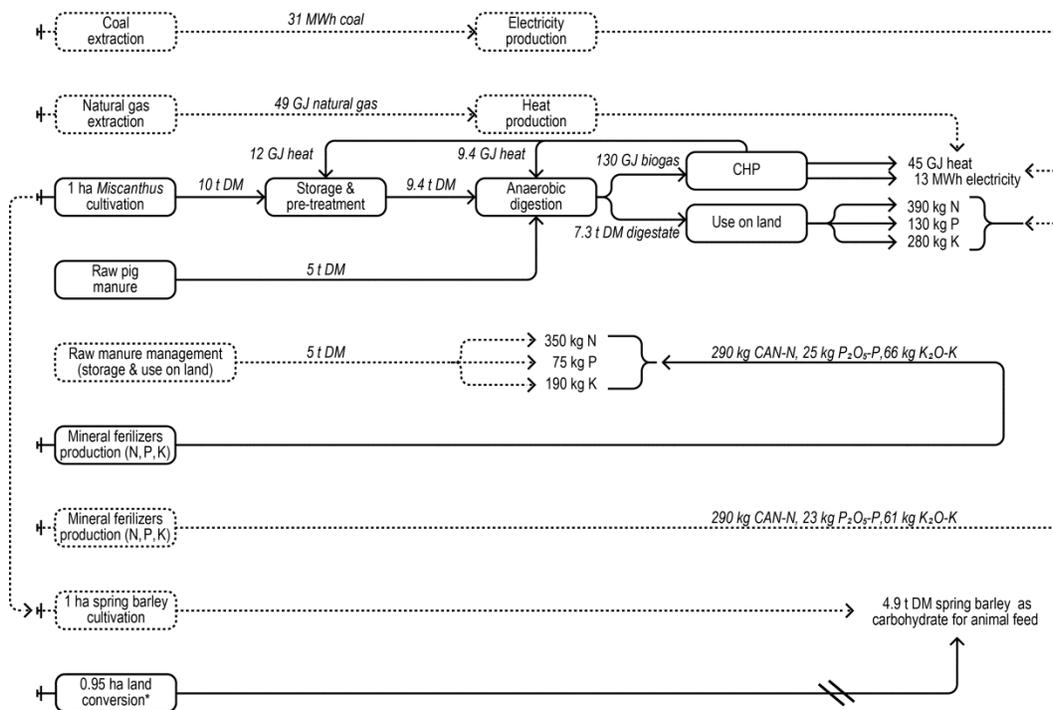


Figure S8. Process flow diagram for anaerobic co-digestion of *Miscanthus* with raw pig manure. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Values are rounded to 2 significant digits.

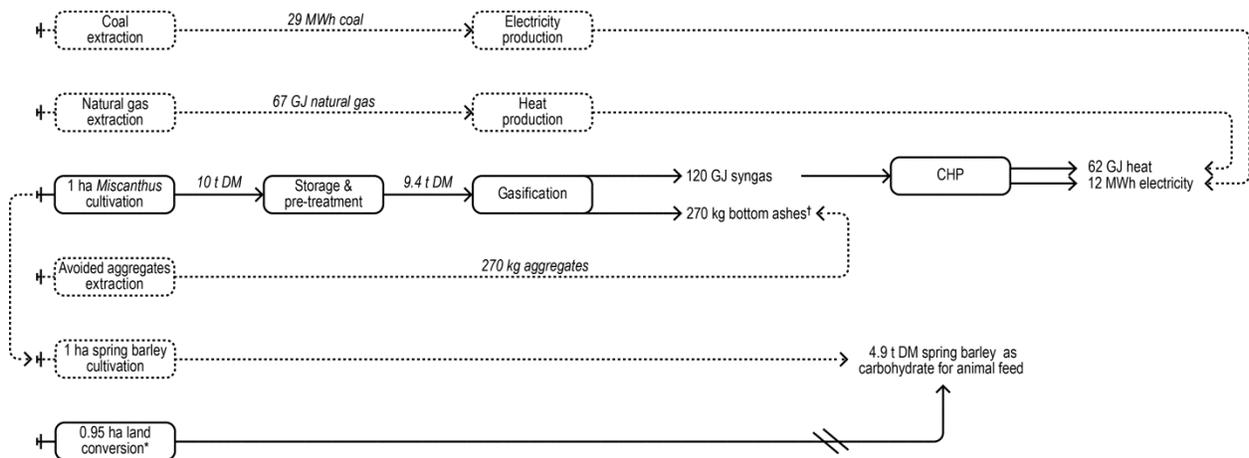


Figure S9. Process flow diagram for gasification of *Miscanthus*. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Fly ashes are considered used as back-filling material in old salt mines, and the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

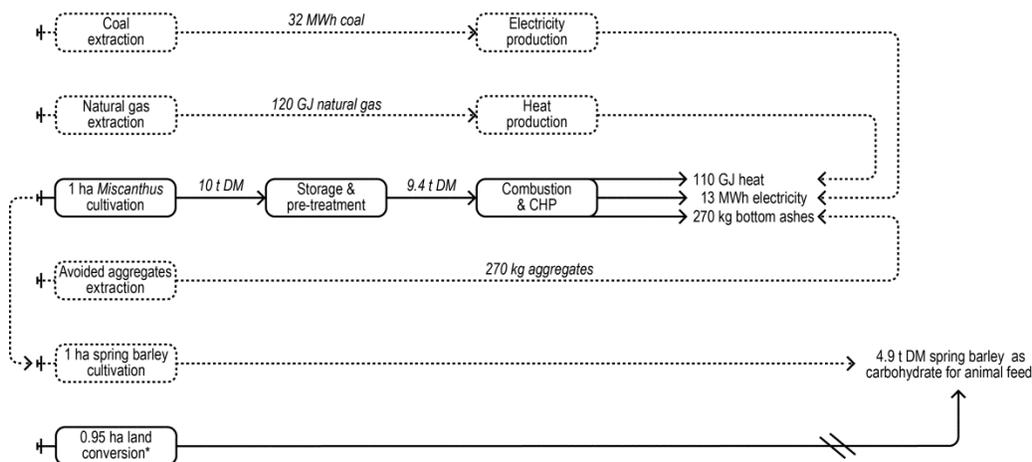


Figure S10. Process flow diagram for combustion of *Miscanthus*. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Values are rounded to 2 significant digits.

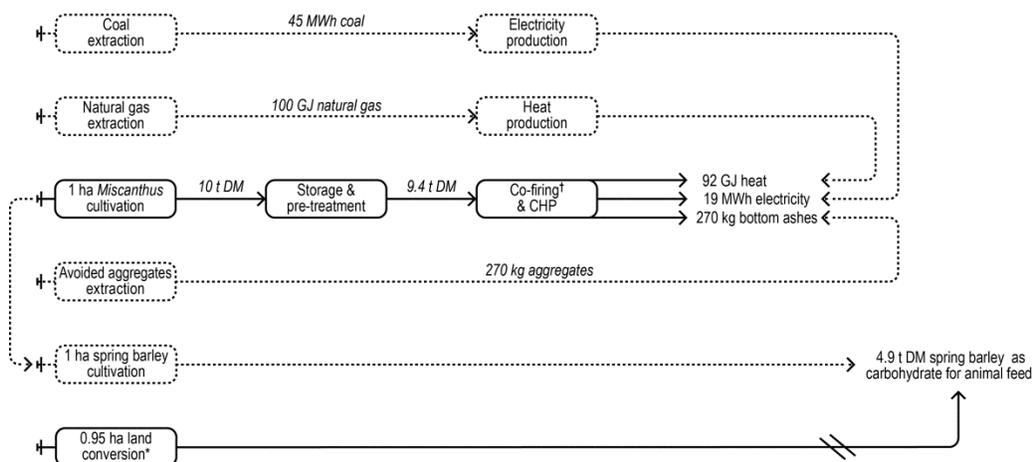


Figure S11. Process flow diagram for co-firing of *Miscanthus*. (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. (†) Based on the data from co-firing Danish plants, the coal that is used here would have 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 CO₂ 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 CO₂ 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 of 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 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); CH₄ pot: methane potential; n.a.: not available.

Parameter	unit	Ryegrass	Willow	<i>Miscanthus</i>
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) ^γ	90 (±6) ^β
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) ^α
P	% DM	0.40 (±0.08) [†]	0.07*	0.49 (±0.08) ^α
K	% DM	0.33 (±0.06) [†]	0.3*	0.69 (±0.2) ^α
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 ^ε	350 ^ε	360 ^ε

* 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 CO₂, CH₄, NH₃, and N₂O 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% ($\pm 4.5\%$) 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 CH₄ and CO₂ caused by dry matter degradation and losses within the storage period were modeled based on the assumed dry matter losses. The CO₂ emission was calculated proportional to the total dry matter loss based on the concentration of carbon in the dry matter, assuming aerobic conditions. The CH₄ 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 N₂O, NH₃ and NO₃ (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. Further, 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. In addition, 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 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% ($\pm 10\%$) of the initial DM content (20, 31). 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). No drying was assumed required for spring harvested *Miscanthus*, due to its low water content (10%) at harvest. Biomasses undergoing anaerobic digestion require size comminution (assumed 10-50 mm); this was considered by including an electricity consumption of $7.5 \text{ kWh t}^{-1} \text{ DM}$ (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 $1.3 \text{ GJ t}^{-1} \text{ DM}$.

3.3.2 Pre-treatments: gasification

The gasification process in fluidized bed typically requires biomass with water content below 20% (35). For the three crops assessed the drying process was assumed as described in 3.3.1 for anaerobic digestion. 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 and comminuted, and homogeneous process conditions are facilitated.

3.3.3 Pre-treatments: combustion and co-firing with fossil fuel

For combustion and co-firing the approach for the drying process was the same as for anaerobic digestion and 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.

BtE	Pre-treatment					Energy conversion			
	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)
Willow	AD	X	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)
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)

* 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 (ca. 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 $450 \text{ Nm}^3 \text{ t}^{-1} \text{ VS}$. The yield was set to 70%, i.e., generating $320 \text{ Nm}^3 \text{ t}^{-1} \text{ VS}$ (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 $360 \text{ Nm}^3 \text{ CH}_4 \text{ t}^{-1} \text{ VS}$ 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 $\text{Nm}^3 \text{CH}_4 \text{t}^{-1} \text{VS}$. 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 ($198\text{-}510 \text{Nm}^3 \text{CH}_4 \text{t}^{-1} \text{VS}$, see Table S4).

Table S3. Composition of ryegrass, willow and *Miscanthus* in terms of lipids, carbohydrates, proteins, lignin and relative calculated methane potential. $\text{CH}_4 \text{pot}$: methane potential ($\text{Nm}^3 \text{t}^{-1} \text{VS}$); %VS (concentration of the parameter (e.g., lipids) as % of VS); D_{raw} : degradability of the raw substrate (% $\text{CH}_4 \text{pot}$); D_{PT} : degradability of the pre-treated substrate (% $\text{CH}_4 \text{pot}$). The values of D_{PT} for willow and *Miscanthus* are based on laboratory batch-tests (33).

Parameter	$\text{CH}_4 \text{pot}$	Ryegrass			Willow			<i>Miscanthus</i>		
		%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		
$\text{CH}_4 \text{pot}$ (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.

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

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

$$W_{\text{biogas,man}} = VS_{\text{man}} \cdot W_{\text{man}} \cdot \text{yield}_{\text{man}} \cdot \rho / (\text{CH}_4 \% \cdot 1000) \quad \text{Eq S3.}$$

$$W_{\text{biogas,crop}} = VS_{\text{man}} \cdot W_{\text{man}} \cdot \text{yield}_{\text{crop}} \cdot \rho / (\text{CH}_4 \% \cdot 1000) \quad \text{Eq S4.}$$

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

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

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)

$VS_{\text{deg,man}}$: 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)

$VS_{\text{deg,crop}}$: VS degraded from the crop after the first digestion stage (kg)

$\text{Yield}_{\text{man}}$: methane yield of the manure after the first digestion stage ($\text{Nm}^3 \text{t}^{-1} \text{VS}$)

$\text{Yield}_{\text{crop}}$: methane yield of the crop after the first digestion stage ($\text{Nm}^3 \text{t}^{-1} \text{VS}$)

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%.

$$\frac{DR_{\text{man}}}{\text{CH}_{4\text{pot man}} \cdot \text{Tot Yield}_{\text{man}}} = \frac{DR_{\text{crop}}}{\text{CH}_{4\text{pot crop}} \cdot \text{Tot Yield}_{\text{crop}}} \quad \text{Eq. S7.}$$

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)

$\text{CH}_{4\text{pot man}}$: methane potential of the manure ($\text{Nm}^3 \text{CH}_4 \text{t}^{-1} \text{VS}$)

$\text{CH}_{4\text{pot crop}}$: methane potential of the crop ($\text{Nm}^3 \text{CH}_4 \text{t}^{-1} \text{VS}$)

$\text{Tot Yield}_{\text{man}}$: total methane yield of the manure (% $\text{CH}_{4\text{pot man}}$)

$\text{Tot Yield}_{\text{crop}}$: total methane yield of the crop (% $\text{CH}_{4\text{pot crop}}$)

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 °C to 37 °C. The fugitive emission of methane was estimated to 1% of the methane produced, based on recent LCA studies (8, 32, 41). Emissions of biogenic CO₂ were estimated as a function of the biogenic CH₄ releases, based on the methodology described by (8). Based on this, the ratio CO₂ to CH₄ 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% ($\pm 4\%$), 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).

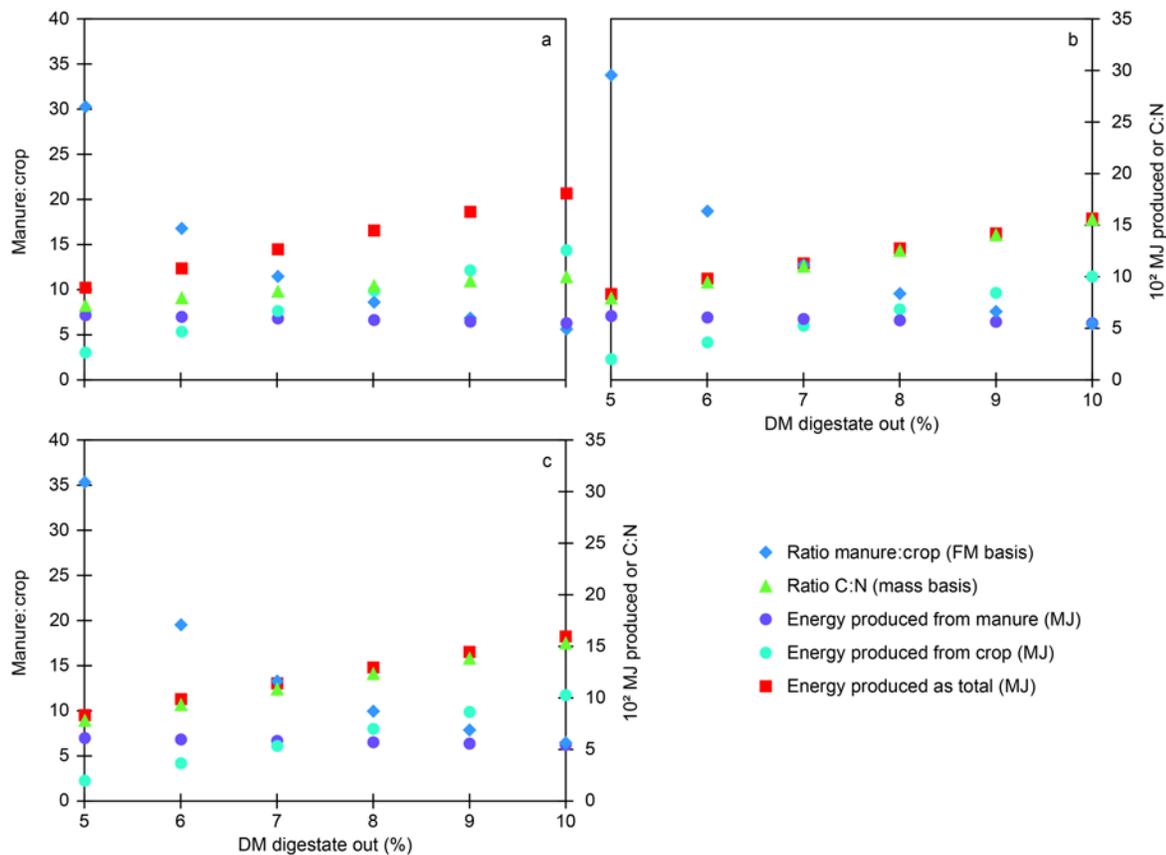


Figure S12. Illustration of I) manure to crop ratio (FM basis) of the mix manure-crop fed into the digestion plant, II) C to N ratio of the mix manure-crop fed into the digestion plant, III) share of the total energy produced from manure (MJ t^{-1} FM input), IV) share of the total energy produced from 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 °C, 35-40 days	(45)*	
	233-327	Lab batch test 35 °C, 28 days	(46)*	
	300-320	Lab/semi-continuous/35 °C/28 days	(47)*	
	320-510 [†]	Lab batch tests/70-80 days	(48)*	290
	310-360	Lab batch tests/35 °C/28 days	(49)*	
	410 [†]	Lab and pilot scale tests	(50)	
	390	Pilot scale	(51)	
Other grasses	361 [†]	Lab batch test	(52)	
	197-470	Review of different grass species	(53)	
Willow	305	Modeling	(54)	
	300	With pretreatment	(33)	240
<i>Miscanthus</i>	90	No pretreatment	(51)	
	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 plant. 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 CH₄, CO and CO₂ and then to CO₂ 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 ₂ O (% 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)
<i>Miscanthus</i> 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 pyrolysis 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% \pm 2%). The associated total efficiency was between 85% and 95% with average 90% (\pm 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 CO₂) 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 along 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 $\pm 3\%$). This was also the average annual net electricity efficiency of *Avedoerevaerket*. The related total efficiency was set to 90% ($\pm 5\%$) 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 ^a
CHP	Assens (1999)	Wood chips, mix (wood waste, residues)	Pneumatic feeders; oscillating grate	25	85 ^β
CHP	Rudkøbing (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/Sakskøbing (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	Avedøreværket (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 [†]	93 [†]

*Gross efficiency.

^a 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 selected 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. Data are from (44).

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 ₂ O	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	μg GJ ⁻¹	4577	8492	12088	2314
NMVOC	g GJ ⁻¹	10	2.3	<0.78	<5.1
ΣPAH	μg 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 CH₄ from digestate storage was calculated using the same approach as for crop storage (section 3.2). The emission of biogenic CO₂ was estimated as for the biogas, i.e., as a function of the biogenic CH₄ 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 N₂O 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 NH₃-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 N₂O emission (i.e., N₂O produced from secondary reactions involving NH₃, NO_x and leached N) were quantified based on IPCC (74). With respect to this, the N₂O flows associated with use on land shown in Figure S16-S18 only refer to the direct N₂O 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 CO₂ was determined based on the findings of (78). Based on this, it was considered that 66% of the initial C applied is emitted as CO₂ 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 by 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; CO₂-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; CO₂-C_{avoided}: avoided carbon emission due to (fossil) energy substitution. Negative values here represent inflows, sinks and avoided emissions (e.g., uptake, ΔSOC, etc.).

Phase	Emission	Ryegrass				Willow				<i>Miscanthus</i>			
		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 ₂ O-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 ₂ O-N(dir.)	9.7	-	-	-	7.9	-	-	-	5.9	-	-	-
	N ₂ O-N(ind.)	3.3	-	-	-	2.4	-	-	-	1.8	-	-	-
	NO ₃ ⁻ -N	290	-	-	-	240	-	-	-	180	-	-	-
	N in soil	270	-	-	-	220	-	-	-	160	-	-	-

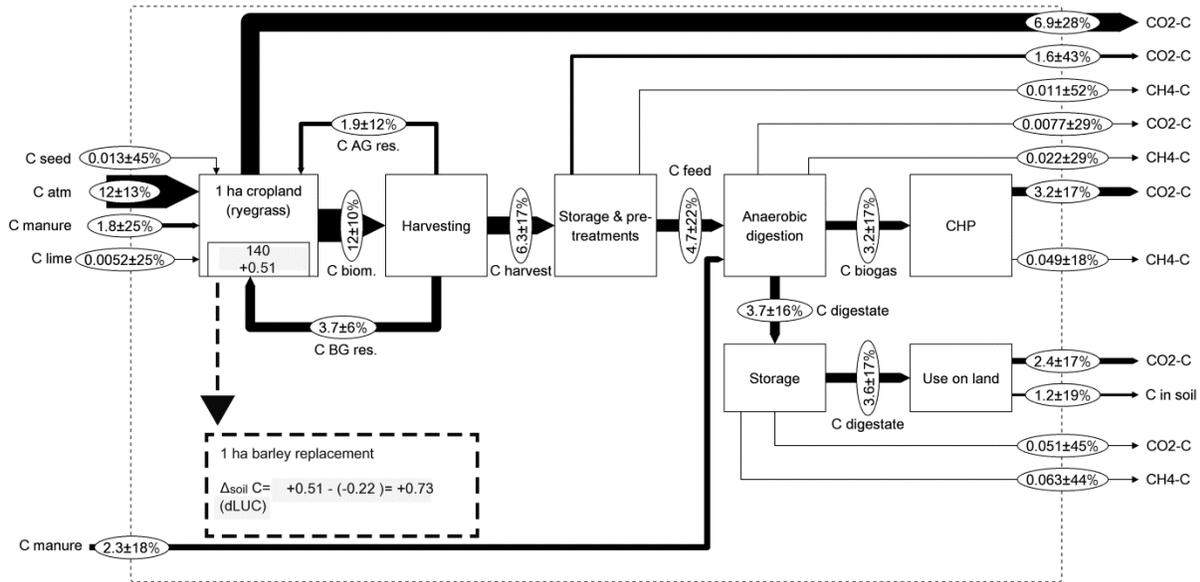


Figure S13. Illustration of the C flows breakdown (t C ha⁻¹ y⁻¹) for anaerobic co-digestion of ryegrass with raw pig manure (values rounded to 2 significant digits). AG stands for above-ground residues and BG stands for below-ground residues. Carbon fossil emissions associated with machinery used in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

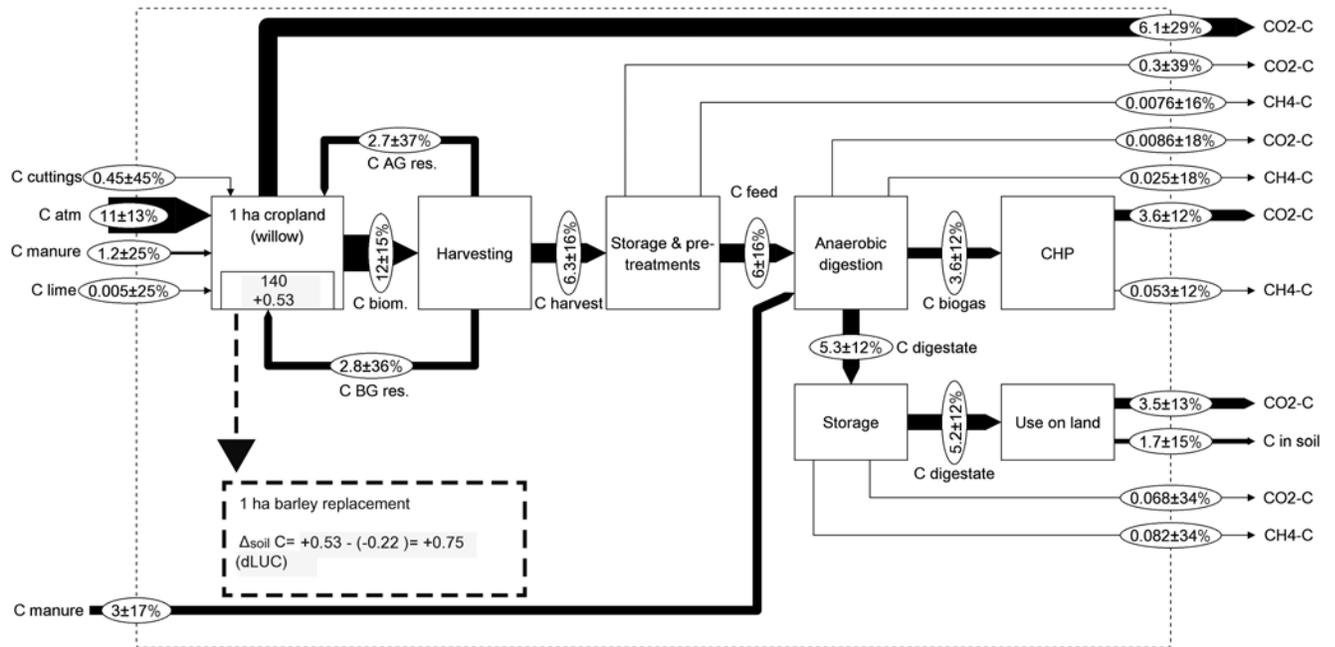


Figure S14. Illustration of the C flows breakdown (t C ha⁻¹ y⁻¹) for anaerobic co-digestion of willow with raw pig manure (values rounded to 2 significant digits).

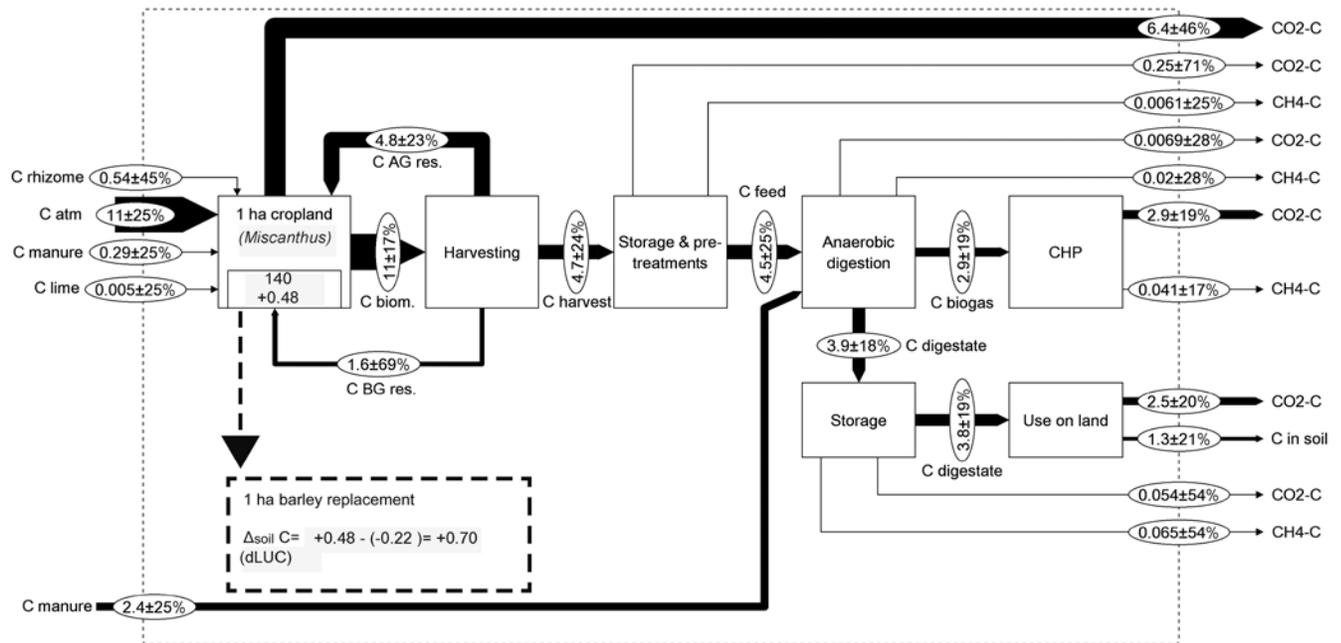


Figure S15. Illustration of the C flows breakdown ($\text{t C ha}^{-1} \text{y}^{-1}$) for anaerobic co-digestion of *Miscanthus* with raw pig manure (values rounded to 2 significant digits).

Note that the data displayed for the cultivation phase of *Miscanthus* and willow refer to the period when the plantation is established (i.e., years 4-20). Additionally, for willow, the data presented are for the years with 100% slurry application only (which occurs at y6, y9, y12, y15 and y18). The detailed LCI of the three crops studied can be found in (9). Note also that inflows and sinks (e.g., uptake, ΔSOC , etc.) are displayed with positive values on the left-side of the Figures (while the same are reported along with a 'minus' in Table S8 to be distinguished from the outflows/emissions which are reported along with a 'plus').

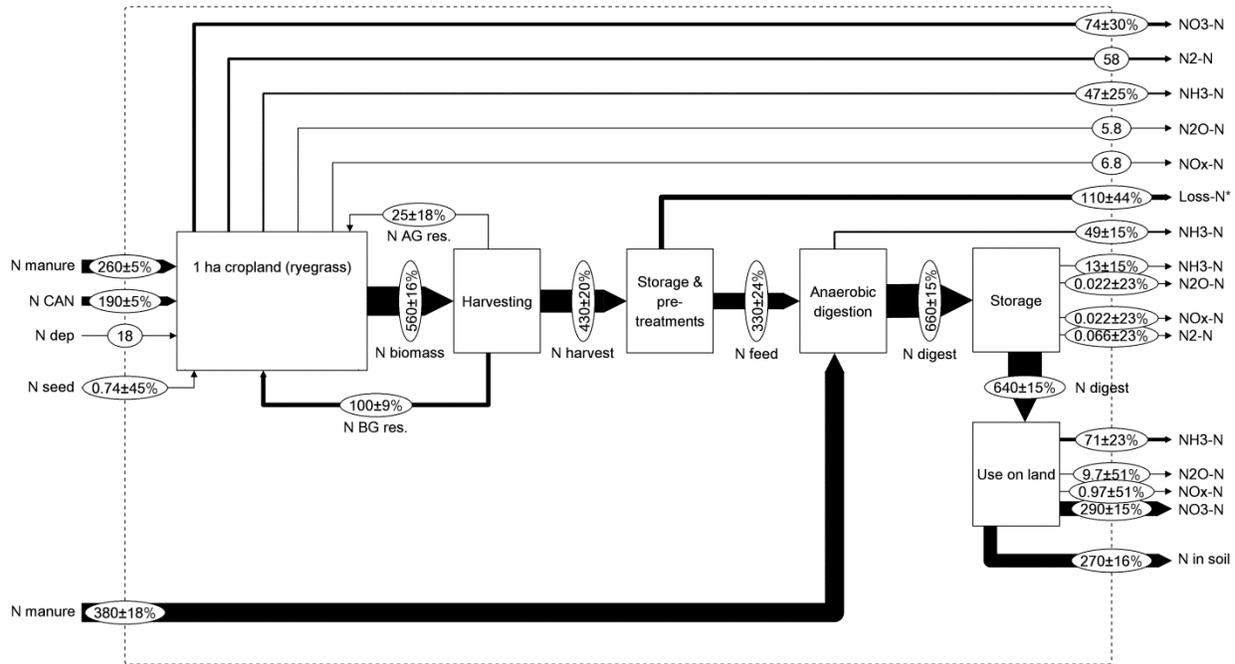


Figure S16. Illustration of the N flows breakdown ($\text{kg N ha}^{-1} \text{y}^{-1}$) for anaerobic co-digestion of ryegrass with raw pig manure (values are rounded to 2 significant digits). AG and BG stands for above- and below- ground residues; N* stands for total unspecified N losses during crop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

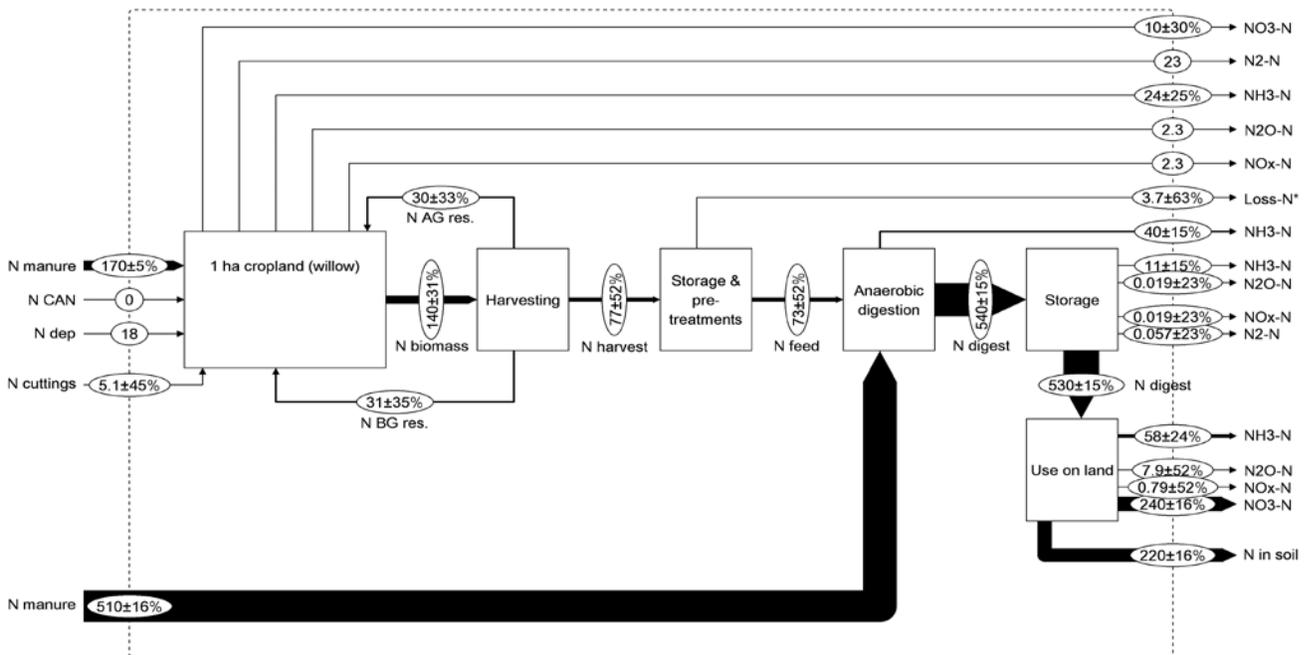


Figure S17. Illustration of the N flows breakdown ($\text{kg N ha}^{-1} \text{y}^{-1}$) for anaerobic co-digestion of willow with raw pig manure (values rounded to 2 significant digits).

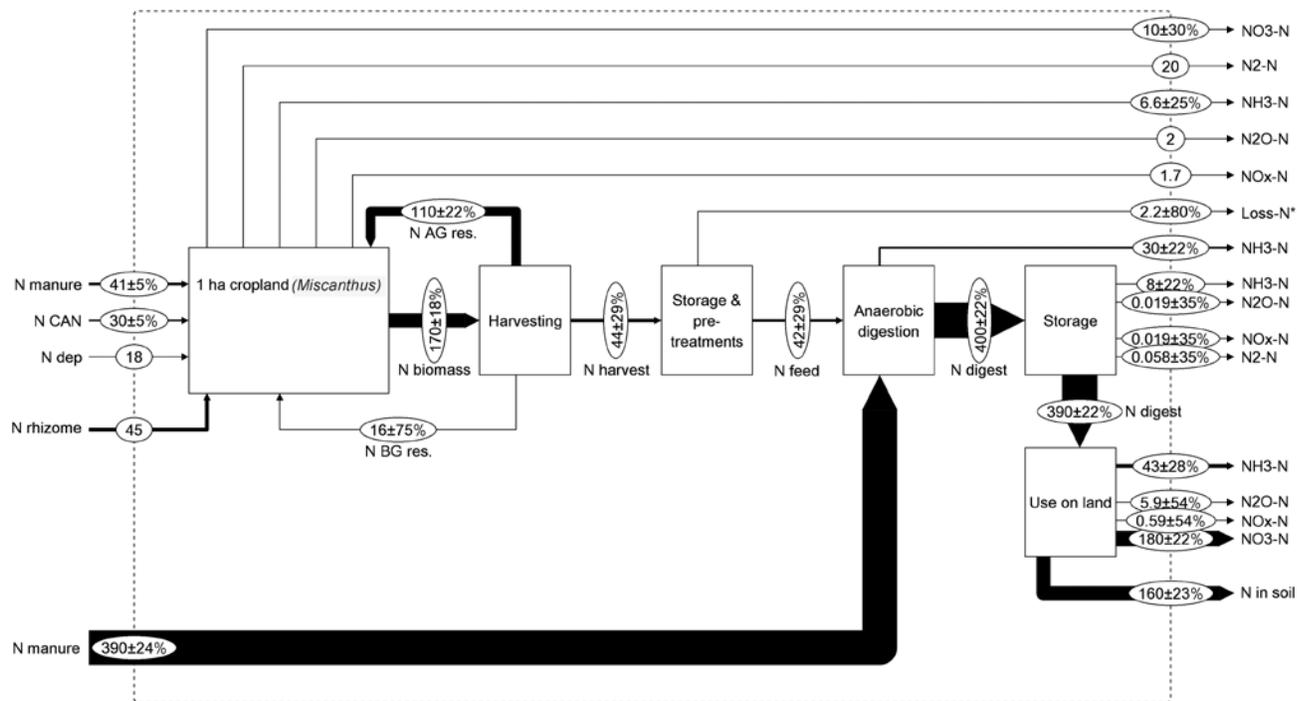


Figure S18. Illustration of the N flows breakdown ($\text{kg N ha}^{-1} \text{y}^{-1}$) for anaerobic co-digestion of *Miscanthus* with raw pig manure (values rounded to 2 significant digits).

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 initially yielded energy per hectare (dry basis). For combustion (CO) and co-firing (CF) the efficiency reported (η_{el}) is a net efficiency (i.e., plant own consumption has been previously subtracted).

		Ryegrass				Willow				<i>Miscanthus</i>			
		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 [†]	4.6 [†]	0.9	0.3	5.1 [†]	5.1 [†]	0.7	0.7	6.1 [†]	6.1 [†]
	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 m²).

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 four 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 (1700)*

* Total value (1658) rounded to two significant digits.

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 CO₂ 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. CO₂-C atm: carbon uptake from atmosphere; CO₂-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; CO₂-C dig: C released after digestate application on land; NH₃-N, N₂O-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 (±0.6)	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	5.5 (±4.5)	4.8 (±1.3)	5.5 (±4.5)	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 ₂ O-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. The results reported in (18) were used as proxy to estimate average value and uncertainty range.

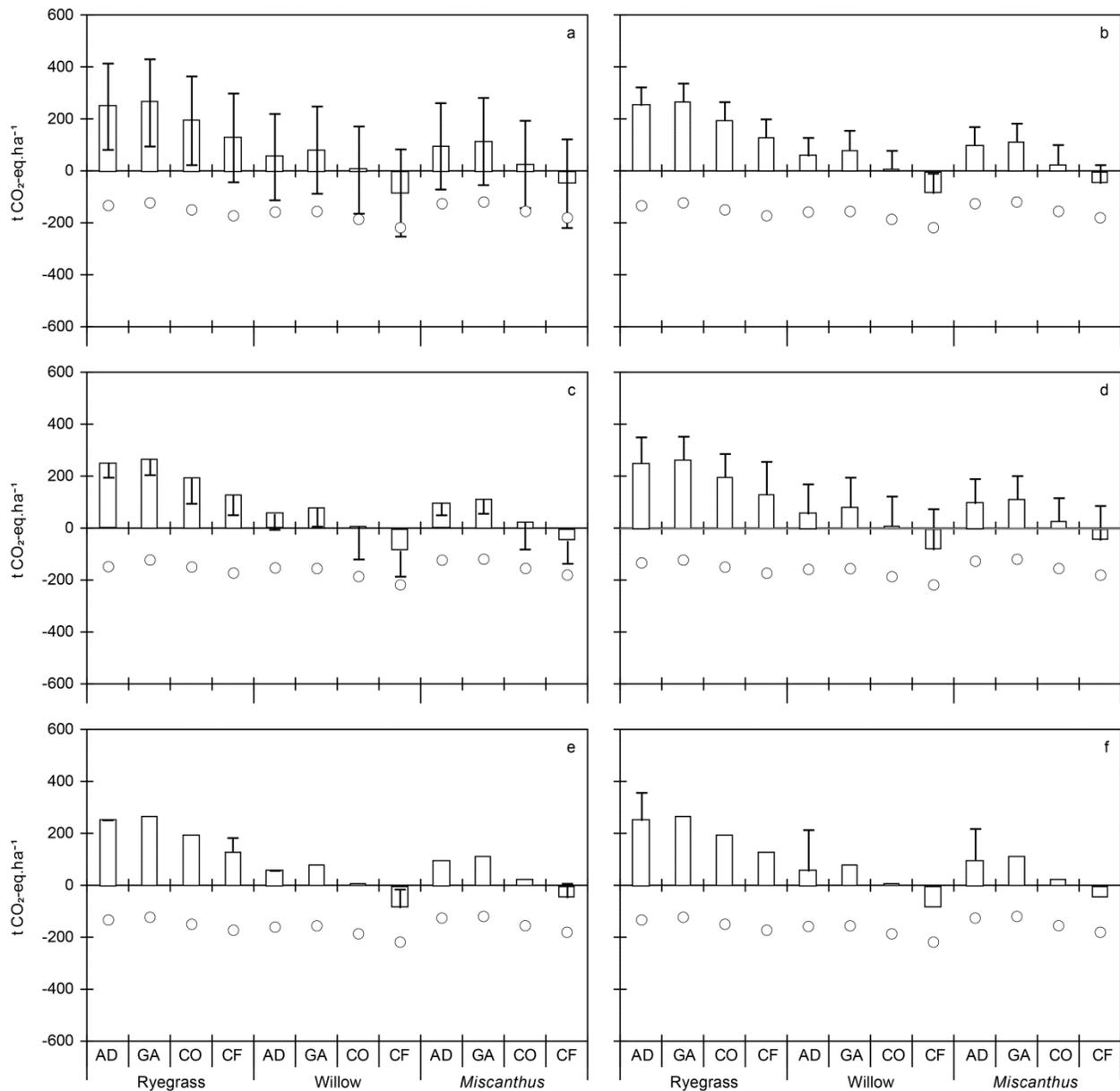


Figure S19. Sensitivity analysis: the error bars illustrate the variation in the LCA results for GW compared with the baseline LCA results. The 'circle' indicates the GW saving corresponding to a 35% GHG reduction compared with the reference (used as comparative measure-stick). The following are displayed: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

$$DL = \text{Yield} \cdot K_D \quad \text{Eq. S8.}$$

$$\text{C loss drying} = DL \cdot C \quad \text{Eq. S9.}$$

$$\text{N loss drying} = DL \cdot N \quad \text{Eq. S10.}$$

$$LC_{dr} = \text{C loss drying} / (\text{yield} \cdot C) \quad \text{Eq. S11.}$$

$$LN_{dr} = \text{N loss drying} / (\text{yield} \cdot N) \quad \text{Eq. S12.}$$

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

$$SL = (\text{Yield} - DL) \cdot K_L \quad \text{Eq. S13.}$$

$$C \text{ loss st} = SL \cdot C \quad \text{Eq. S14.}$$

$$N \text{ loss st} = SL \cdot N \quad \text{Eq. S15.}$$

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

$$CO_2 = C \text{ loss st} - (CH_4 \cdot 12/16) \cdot 44/12 \quad \text{Eq. S17.}$$

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

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

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)

CH₄: emission of methane during storage (t CH₄ ha⁻¹ y⁻¹)

CO₂: emission of carbon dioxide during storage (t CO₂ 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)

CH₄_{pot}: methane potential (Nm³ CH₄ t⁻¹ DM)

MCF: methane conversion factor (% CH₄_{pot})

K: ratio CO₂/CH₄ 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 m³ CH₄ to kg CH₄ (CH₄ density at 20°C). The ratio 12/16 is the conversion factor between methane and carbon emissions (i.e., kg C kg⁻¹ CH₄). 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:

$$El = \text{Crop fed} \cdot VS \cdot CH_{4\text{pot}} \cdot CH_{4\text{yield}} \cdot LHV_{CH_4} \cdot \eta_{el} / 3.6 \quad \text{Eq. S20.}$$

$$Ht = \text{Crop fed} \cdot VS \cdot CH_{4\text{pot}} \cdot CH_{4\text{yield}} \cdot LHV_{CH_4} \cdot \eta_{th} \quad \text{Eq. S21.}$$

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

Gasification:

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

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

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

Combustion and co-firing:

$$El = \text{Crop fed} \cdot LHV_{wb} \cdot \eta_{el} / 3.6 \quad \text{Eq. S26.}$$

$$El = \text{Crop fed} \cdot LHV_{wb} \cdot \eta_{th} \quad \text{Eq. S27.}$$

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

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 (Nm³ CH₄ t⁻¹ DM)

CH_{4 pot}: methane yield (% CH_{4 pot})

LHV_{CH4}: lower heating value of methane (STP) (35.2 MJ Nm⁻¹ CH₄)

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:

$$\text{CO}_2 = [\text{Yield} \cdot \text{C} \cdot (1 - \text{LC}_{\text{dr}} - \text{LC}_{\text{st}}) \cdot \text{CC} - E_{\text{C,f}} - E_{\text{CCH}_4}] \cdot 44/12 \quad \text{Eq. S29.}$$

$$\text{CC} = \frac{\text{VS} \cdot \text{CH}_{4\text{pot}} \cdot \text{CH}_{4\text{yield}}}{(\% \text{CH}_4 \cdot V_m)} \cdot \frac{1}{10^3} \cdot \frac{C_m}{C} \quad \text{Eq. S30.}$$

Gasification:

$$\text{CO}_2 = [\text{Yield} \cdot \text{C} \cdot (1 - \text{LC}_{\text{dr}} - \text{LC}_{\text{st}}) \cdot \text{CCE} - E_{\text{CCH}_4}] \cdot 44/12 \quad \text{Eq. S31.}$$

Combustion and co-firing:

$$\text{CO}_2 = [\text{Yield} \cdot \text{C} \cdot (1 - \text{LC}_{\text{dr}} - \text{LC}_{\text{st}}) - E_{\text{CCH}_4}] \cdot 44/12 \quad \text{Eq. S32.}$$

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_{CCH_4} : emission of unburned methane (t C ha⁻¹ y⁻¹)

The ratio 44/12 is the conversion factor between CO₂ and C emissions (i.e., kg CO₂ 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_{CCH_4}) can be recalculated from Table S8.

Emissions from digestate storage

$$CH_4 = \text{Yield}_{\text{dig}} \cdot CH_{4\text{pot}} \cdot 0.67 \cdot \text{MCF} \quad \text{Eq. S33.}$$

$$CO_2 = CH_4 \cdot K \quad \text{Eq. S34.}$$

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

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

$$N \text{ loss dig} = C \text{ loss dig}/C \cdot N \quad \text{Eq. S37.}$$

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

Where:

CH_4 : emission of methane (t CH₄ ha⁻¹ y⁻¹)

CO₂: emission of carbon dioxide (t CO₂ 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 (Nm³ CH₄ t⁻¹ DM)

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

K: ratio CO₂/CH₄ 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

$$\text{CO}_2 = \text{Yield} \cdot \text{C} \cdot (1 - \text{CC} - \text{LC}_{\text{dr}} - \text{LC}_{\text{st}} - \text{LC}_{\text{st dig}}) \cdot 0.74 \cdot 44/12 \quad \text{Eq. S39.}$$

$$\text{NH}_3 = \text{Yield} \cdot \text{N} \cdot (1 - \text{NC} - \text{LN}_{\text{dr}} - \text{LN}_{\text{st}} - \text{LN}_{\text{st dig}}) \cdot 0.11 \quad \text{Eq. S40.}$$

$$\text{NO}_3 = \text{Yield} \cdot \text{N} \cdot (1 - \text{NC} - \text{LN}_{\text{dr}} - \text{LN}_{\text{st}} - \text{LN}_{\text{st dig}}) \cdot 0.45 \quad \text{Eq. S41.}$$

$$\text{N}_2\text{O direct} = \text{Yield} \cdot \text{N} \cdot (1 - \text{NC} - \text{LN}_{\text{dr}} - \text{LN}_{\text{st}} - \text{LN}_{\text{st dig}}) \cdot 0.015 \quad \text{Eq. S42.}$$

$$\text{NO}_x = \text{N}_2\text{O direct} \cdot 0.1 \quad \text{Eq. S43.}$$

$$\text{N}_2\text{O indirect} = \text{N leached} \cdot 0.0075 + (\text{NH}_3 + \text{NO}_x) \cdot 0.01 \quad \text{Eq. S44.}$$

Where:

CO_2 : carbon dioxide produced (t $\text{CO}_2 \text{ ha}^{-1}$ in 20y)

NH_3 : ammonia emission (t N $\text{ha}^{-1} \text{ y}^{-1}$)

NO_3 : nitrates leaching (t N $\text{ha}^{-1} \text{ y}^{-1}$)

NO_x : NO_x emission (t N $\text{ha}^{-1} \text{ y}^{-1}$)

N_2O : direct: nitrous oxide emission (direct) (t N $\text{ha}^{-1} \text{ y}^{-1}$)

N_2O : indirect: nitrous oxide emission (indirect) (t N $\text{ha}^{-1} \text{ y}^{-1}$)

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)

$\text{LC}_{\text{st dig}}$: 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)

$\text{LN}_{\text{st dig}}$: 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

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

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

Where:

$\text{GHGs}_{\text{EU 35\%}}$: GHGs emission (of the individual bioenergy scenario under assessment) that should be achieved to fulfill the EU directive target (t CO₂-eq. ha⁻¹)

$\text{GHGs}_{\text{EU 35\% relative}}$: 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 CO₂-eq. ha⁻¹)

$\text{GHGs}_{\text{fossil ref}}$: GHGs emission of the reference fossil fuel system where the hectare of land is used for spring barley cultivation (t CO₂-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 CO₂ 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 N₂O 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 CO₂ 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 CO₂ 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 CO₂ emission (310 000 kg CO₂eq. 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 (<i>Miscanthus</i> , 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 ₂ O (direct) ^b	N ₂ O (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 year 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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Appendix C. Supporting Information for Paper IV

Supporting Information for:

Environmental Consequences of Future Biogas Technologies Based on Separated Slurry

Lorie Hamelin, Marianne Wesnæs, Henrik Wenzel, Bjørn M. Petersen

54 pages, 2 figures, 32 tables

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1. Scope

The technological scope for biogas production is based on the best technologies available in Denmark.

This includes:

- A short (less than 7 days) storage time of the solid fraction before it is used as an input for biogas production;
- A two-steps biogas production, operating at mesophilic temperatures, where the post-digestion tank is covered with an air-tight cover. This means that most of the leftover methane (CH₄) that has not been captured during the first digestion is recuperated and that the overall biogas system is run without uncontrolled gaseous emissions. The first digestion step is considered to yield 90 % of the final biogas yield;
- A biogas engine with high efficiency: 46 % for heat and 40 % for electricity with a total efficiency of 86 %;
- A covered storage of all slurry and fractions prior and after the digestion.

2. Functional Unit

The functional unit was defined as “the management of 1 ton of post-animal slurry”. In this system, where biogas alternatives from separated slurry are compared, this is the obvious service provided to society. The production of energy (electricity and/or heat) was discarded as a functional unit for several reasons. First, slurry biogas is produced from animal slurry, a co-product from another activity, namely animal production. Therefore, the production of slurry is not going to increase as a result of an increased demand for heat and power based on slurry biogas, so if an environmental assessment is to be made on energy producing technologies, it should rather include these energy technologies that can react to a demand change, which would be more relevant for policymaking. Moreover, in this case, the references for producing energy involve e.g. coal, natural gas, etc., and these would need to be included as well to

provide a fair comparison, if providing energy was the main objective. Rather, the actual service provided to society is the management of slurry, and what is relevant for policymakers is an environmental assessment of different ways of dealing with this produced slurry. This is why the reference scenario consists of the management of slurry where slurry is used as a fertilizer without further processing.

3. Preconditions for the Reference Scenario

The biogas alternatives include both fattening pig and dairy cow slurry. Accordingly, two reference scenarios are defined: one assessing the life-cycle flow of pig slurry (REF-pig) and one assessing the life-cycle flow of dairy cow slurry (REF-cow). In order to define these reference scenarios, it has been necessary to define some preconditions regarding e.g. housing units, type of storage, technology for application to the field and a reference cropping scenario. The main preconditions that needed to be defined are described below:

For fattening pigs, these pre-conditions include:

Housing system: A housing system with fully slatted floors has been chosen due to the fact that fully slatted floor was the most common housing system for fattening pigs in Denmark in 2006-2007 (approximately half of the housing systems for fattening pigs), according to a personal communication with Hanne Damgaard Poulsen, Faculty of Agricultural Sciences, Aarhus University, October 2008. This assumption was necessary because the reference used for determining the slurry composition distinguishes between the floor systems. A storage time in the pit underneath the animals of approximately 14 days is assumed (1).

Outdoor storage: In Denmark, it is required by law to cover outdoor slurry storage tank in order to reduce ammonia emissions and odor. For the reference scenario, the considered cover consists of a floating layer of straw as this is the minimum requirement in the law and as this is the cheapest and most widespread method (2).

Crop rotation: Crops are not included in the system boundary; however, a reference crop rotation had to be defined as the flow of applied slurry nutrients (e.g. uptake by crops, leaching to soil and water)

depends on the crops. The crops also determined the amount of slurry and mineral fertilizers to be applied. Based on the representative farm types established by Dalgaard et al. (3) as well as on the Danish guidelines for fertilization (4), a 6 years crop rotation was defined for fields receiving the pig slurry, with slurry N ($\text{kg ha}^{-1} \text{y}^{-1}$) applied indicated in parenthesis: winter barley (133.5), winter rape (133.5), winter wheat (133.5), winter wheat (133.5), spring barley with catch crop (165), spring barley (145).

For dairy cows, the pre-conditions include:

Housing system: The housing conditions are based on a “Cubicle housing system with slatted floor (1.2 m channel)”, these being the most common housing system for dairy cows in Denmark in 2006-2007 (personal communication with Hanne Damgaard Poulsen, Faculty of Agricultural Sciences, Aarhus University, October 2008). As in the case of the pig slurry reference, this assumption was necessary in order to establish the slurry composition. A storage time in the pit of approximately 14 days is assumed (1).

Outdoor storage: As for pig slurry, it is assumed that the cattle slurry is stored outdoor in a concrete slurry tank. When storing cattle slurry, a natural crust, or floating layer, will be formed due to the fibrous material contained in the slurry (5-6). In Denmark, this is regarded as a sufficient cover (2,7). Accordingly, the reference scenario considers that the cover consists of only this organic matter based natural crust without the addition of any other floating materials (e.g. straw, leca pebbles, permeable membrane, etc.).

Crop rotation: As for the pig slurry, a crop rotation had to be defined in order to assess the fate of slurry nutrients as well as to determine the amount of slurry and mineral fertilizers to be applied. Based on the representative farm types established by Dalgaard et al. (3) as well as on the Danish guidelines for fertilization (4), a 5 years crop rotation was defined for fields receiving the cow slurry, with slurry N ($\text{kg ha}^{-1} \text{y}^{-1}$) applied indicated in parenthesis: spring barley harvested as whole crop silage (156), grass clover mixture (182), grass clover mixture (182), spring barley with catch crop (0), spring barley (132).

Common to both fattening pigs and dairy cows, the following preconditions have been considered:

Pre-tank: In connection with the housing units is a pre-tank from which the slurry is pumped to the outdoor storage.

Transport distance from storage to field: Based on different Danish studies (8-9), the average transport distance for farmers applying the slurry to their own fields is about 5 km and below. For such small distances, it is common to use a tractor with trailer. However, if the transport of slurry to the fields is more than 10 km, transport by truck is required by law. Therefore, a transport distance of 10 km has been used for the reference.

Slurry spreading: According to (10), 68 % of all slurry was spread by trail hose tanker in Denmark (in 2004), and this is still the most common method today (personal communication with Thorkild Birkmose, Landscentret, Dansk Landbrugsrådgivning, October 2008). Therefore, it is considered that slurry is applied with trail hose tankers to the field in the reference scenario. It is assumed that slurry is applied to all crops in the crop rotation pattern, with a farm average of 140 kg N ha⁻¹ y⁻¹. It is also assumed that the slurry is applied during spring.

Soil types: Relevant soil types for pig and cow production in Denmark includes both clay and sandy soils (11-12, 3). Accordingly, both soil types are taken into account; in the present paper, sand is considered for the main scenario and as a sensitivity analysis, the assessment is performed using clay soil.

4. Reference Slurry Composition

The reference slurry composition was determined based on the Danish normative system for assessing slurry composition (13-14), and based on mass balances accounting for all input and output to the slurry flow. It is necessary to set this reference slurry composition for the purpose of this LCA since it is the basis for all subsequent emission flows. However, it is recognized that the composition of slurry is, in practice, seldom “standard”, varying upon diets, management practices, animal age and type, temperature, etc.

Table S1 presents, for both pig and cow slurry, the entire composition of the reference slurry considered in the study, for the three main life cycle stages of the slurry, i.e. post-animal, post-housing

(as it leaves the temporal in-house storage) and post-storage (as it leaves the outdoor storage). This table also details the references and assumptions used in establishing these reference slurries. Table S2 shows the assumptions used for estimating the N losses occurring between the different slurry stages, which were necessary for establishing the slurry N composition. Values in Table S2 apply for both pig and cow slurry, unless otherwise specified.

Table S1. Reference slurries composition

Parameter	Slurry type	Slurry post-animal ^a	Slurry post-housing ^b	Slurry post-storage ^{c,d}	Source and assumptions
Total N (kg ton ⁻¹)	Pig	6.60	5.48	4.80	N post-animal from (14). Losses considered (during housing and during storage): NH ₃ , N ₂ O, N ₂ , NO. See Table S2 for details about N losses. For cow, the N from straw addition in-house is estimated as 0.0913 kg N per ton slurry post-animal ^e .
	Cow	6.87	6.34	5.79	
P (kg ton ⁻¹)	Pig	1.13	1.13	1.04	P post-animal from (14). No losses considered during housing and storage. For cow, the P from straw addition in-house is estimated as 0.0124 kg P per ton slurry post-animal ^e .
	Cow	1.02	1.03	0.98	
K (kg ton ⁻¹)	Pig	2.85	2.85	2.60	K post-animal from (14). No losses considered during housing and storage. For cow, the K from straw addition in-house is estimated as 0.269 kg K per ton slurry post-animal ^e .
	Cow	5.81	5.90	5.65	
DM (kg ton ⁻¹)	Pig	77.4	69.7	61.0	DM post-storage from (14). Losses during storage: 5 % of the post-housing values; losses during housing: 10 % of the post-animal value. Assumptions for losses during storage and housing based on (13).
	Cow	125.7	113.2	103.0	
VS (kg ton ⁻¹)	Pig	64.2	56.5	48.8	VS are assumed to constitute 80 % of the DM content of any slurry types. Losses considered during storage and housing (absolute values) are the same as for DM (i.e. it is assumed that all DM lost was VS).
	Cow	104.2	91.7	82.4	
C (kg ton ⁻¹)	Pig	37.0	33.3	29.2	C post-storage = 47.9 % of DM post-storage for pigs, and 43.9 % of DM post-storage for cows. Estimates based on the ratio C: DM obtained by (15). Losses of C during storage and housing assumed to follow the same pattern as DM (i.e. 5 % of the post-housing values and 10% of the post-animal values, respectively).
	Cow	55.2	49.7	45.2	
Cu (g ton ⁻¹)	Pig	30.0	30.0	27.6	Cu post-storage = 0.0453 % of DM post-storage for pigs, and 0.0113 % of DM post-storage for cows. Estimates based on the ratio Cu: DM obtained by (15). No losses considered during housing and storage.
	Cow	12.1	12.1	11.6	
Zn (g ton ⁻¹)	Pig	89.4	89.4	82.4	Zn post-storage = 0.135 % of DM post-storage for pigs, and 0.0217 % of DM post-storage for cows. Estimates based on the ratio Zn: DM obtained by (15). No losses considered during housing and storage.
	Cow	23.4	23.4	22.4	

^a All values of this column are expressed per ton slurry post-animal. ^b All values of this column are expressed per ton slurry post-housing. ^c All values of this column are expressed per ton slurry post-storage. ^d Post-storage values considers a water addition of 86 kg during storage of pig slurry and of 44 kg during storage of cow slurry. ^e The N, P and K addition from straw added in the stable considers, based on (13), an addition of 1.2 kg of straw per animal per day, a straw DM content of 85 % and a

production of 20400 kg slurry per dairy cow per year. The N, P and K content of straw per kg of DM is 0.005 kg, 0.00068 kg and 0.01475 kg, respectively, based on (13).

Table S2. Assumptions for N losses in the establishment of the reference slurries composition

Losses in-house (kg)	
NH ₃ -N	16 % of N post-animal (pig slurry) and 8 % of N post-animal (cow slurry) (13)
N ₂ O-N	0.002 kg N ₂ O-N per kg N post-animal (16)
N ₂ -N	Assumption that N ₂ -N = N ₂ O-N × 3 (based on data from Dämmgen and Hutchings, (17))
NO-N	Assumption that N ₂ -N = N ₂ O-N × 1 (based on data from Dämmgen and Hutchings, (17))
Losses during storage (kg)	
NH ₃ -N	2 % of N post-housing (13), the N post-housing being estimated according to Poulsen et al. (13), i.e. : N post-animal minus NH ₃ -N losses in-house (and not accounting for other losses).
N ₂ O-N	0.005 kg N ₂ O-N per kg N post-animal (16)
N ₂ -N	Assumption that N ₂ -N = N ₂ O-N × 3 (based on data from Dämmgen and Hutchings, (17))
NO-N	Assumption that N ₂ -N = N ₂ O-N × 1 (based on data from Dämmgen and Hutchings, (17))

5. Alternatives Scenarios: Technology Description and Mass Balances

5.1 Alternative P1

The decanter centrifuge considered for the first slurry separation in this alternative is based on a technology manufactured by GEA Westfalia (18) model UCD 305. The share of the slurry dry matter (DM) and nutrients going to the solid fraction, also referred to as separation efficiencies, was defined based on data from the technology provider except for carbon (C), copper (Cu) and zinc (Zn), for which there were no data. For C, it was assumed that the separation efficiency is the same as for DM. For Cu and Zn, separation efficiencies given in a recent study of Møller et al. (19) were used (centrifuge, pig slurry no.1). Since no polymer addition is involved in the study performed by Møller et al. (19), these efficiencies may be lower as those involved in the actual study, but it is yet a better approximation than simply ignoring Cu and Zn for the rest of the analysis.

Table S3 presents the separation efficiency considered for this separation technology, as well as the mass balances allowing to determine the composition of the separated liquid and solid fractions (for the first separation). Minor inconsistencies may occur in this table due to rounding. The original calculations have been performed with all the decimals. The mass balances for the second separation are presented in section 10.

Table S3. Mass balance for the first separation in Alternative P1 (decanter centrifuge with PAM)

	Amount in slurry before separation	Separation efficiency	<i>Mass balance: amount transferred to the solid fraction (SF)</i>	<i>Mass balance: amount transferred to the liquid fraction (LF)</i>	Solid fraction (SF) composition ^a	Liquid fraction (LF) composition ^b
Unit	kg ton ⁻¹ post housing	%	kg ton ⁻¹ post housing	kg ton ⁻¹ post housing	kg ton ⁻¹ solid fraction	kg ton ⁻¹ liquid fraction
Total mass	1000	22.9	229	771.4	1000	1000
Dry matter (DM)	69.7	87.2	60.8	8.9	265.9	11.6
Total nitrogen (N)	5.48	41.9	2.3	3.2	10.0	4.1
Phosphorus (P)	1.13	90.0	1.0	0.1	4.4	0.1
Potassium (K)	2.85	14.2	0.4	2.4	1.8	3.2
Carbon (C)	33.3	87.2	29.0	4.3	127.1	5.5
Copper (Cu)	0.03	36.2	0.01	0.2	0.05	0.02
Zinc (Zn)	0.09	42.2	0.04	0.05	0.2	0.07

^a Calculated as: (amount transferred to the solid fraction × 1000 kg ton⁻¹) / mass amount transferred to the solid fraction. ^b Calculated as: (amount transferred to the liquid fraction × 1000 kg ton⁻¹) / mass amount transferred to the liquid fraction.

5.2 Alternative P2

The separation technology for Alternative P2 consists of a screw press that was manufactured by Samson Bimatech (20). As in Alternative P1, the separation efficiencies data were defined based on data from the technology provider except for C, for which there were no data, so it has been assumed that the separation efficiency is the same as for DM. Similarly, no data were available from the technology

provider for Cu and Zn, so the separation efficiency for these was based on (19). Table S4 presents the separation efficiency considered for this separation technology, as well as the mass balances allowing to determine the composition of the separated liquid and solid fractions. Minor inconsistencies may occur in this table due to rounding. The original calculations have been performed with all the decimals.

Table S4. Mass balance for pig slurry separation in Alternative P2 (screw press)

	Amount in slurry before separation	Separation efficiency	<i>Mass balance: amount transferred to the solid fraction (SF)</i>	<i>Mass balance: amount transferred to the liquid fraction (LF)</i>	Solid fraction (SF) composition ^a	Liquid fraction (LF) composition ^b
Unit	kg ton ⁻¹ post housing	%	kg ton ⁻¹ post housing	kg ton ⁻¹ post housing	kg ton ⁻¹ solid fraction	kg ton ⁻¹ liquid fraction
Total mass	1000	5.2	52.0	948.0	1000	1000
Dry matter (DM)	69.7	29.6	20.6	49.0	396.9	51.8
Total nitrogen (N)	5.48	6.8	0.4	5.1	7.2	5.4
Phosphorus (P)	1.13	9.1	0.1	1.0	2.0	1.1
Potassium (K)	2.85	2.9	0.08	2.8	1.6	2.9
Carbon (C)	33.3	29.6	9.9	189.7	189.7	24.7
Copper (Cu)	0.03	6.4	1.4	26.5	26.5	30.2
Zinc (Zn)	0.09	6.3	5.6	108.4	108.4	88.4

^a Calculated as: (amount transferred to the solid fraction × 1000 kg ton⁻¹) / mass amount transferred to the solid fraction. ^b Calculated as: (amount transferred to the liquid fraction × 1000 kg ton⁻¹) / mass amount transferred to the liquid fraction.

5.3 Alternative P3

In this alternative, the slurry is separated with the same technology as in Alternative P2. Then, the solid fraction is dried in a tumble dryer and pressed into pellets. Table S5 presents the mass balances performed to estimate the composition of the fibre pellets. The DM content of the fibre pellets, i.e. 88.93 %, was provided by the technology manufacturer, Samson Bimatech (20). Based on that, it was possible to calculate the mass loss occurring during the process (to convert the solid fraction to fibre pellets). It

has been assumed that no losses of nutrient occur during the process, except for N. Losses of N have been calculated based on fibre pellets N measurements from the technology provider, which amounts to a content of 11.59 kg N per ton fibre pellets. The N loss was calculated as the difference between the N from the fibre fraction and the N in the pellets.

Minor inconsistencies may occur in Table S5 due to rounding. The original calculations have been performed with all the decimals.

Table S5. Mass balance for pig slurry separation in Alternative P3 (screw press and pellets fabrication)

Unit	Amount in slurry before separation	Mass balance: amount transferred to solid fraction (SF) (Table S4)	Mass balance: amount in fibre pellets (FP) after the process	Fibre pellets (FP) composition ^a
	kg ton ⁻¹ post housing	kg ton ⁻¹ post housing	kg ton ⁻¹ post housing	kg ton ⁻¹ fibre pellets
Total mass	1000	52.0	23.2 ^b	1000
Dry matter (DM)	69.7	20.6	20.6	889.3
Total nitrogen (N)	5.48	0.4	0.3 ^c	11.75
Phosphorus (P)	1.13	0.1	0.1	4.4
Potassium (K)	2.85	0.08	0.08	3.6
Carbon (C)	33.3	9.9	9.9	424.9
Copper (Cu)	0.03	1.4	1.4	0.06
Zinc (Zn)	0.09	5.6	5.6	0.2

^a Calculated as: (amount in fibre pellets after the process × 1000 kg ton⁻¹) / mass amount in the fibre pellets after the process. ^b Based on the knowledge of the DM content of the FP, i.e. 88.9 %, and on the data from Table S4, this can be calculated as: (396.9 kg DM ton⁻¹ SF × 51.98 kg SF ton⁻¹ slurry post-housing) / (1000 kg ton⁻¹ × 0.889 kg DM kg⁻¹ FP). ^c Based on the technology provider, the fibre pellets should contain 11.59 kg N per ton of fibre pellets. As there is 23.2 kg FP ton⁻¹ slurry post-housing, this corresponds to 0.3 kg N per ton slurry post-housing. The SF contains 0.4 kg N per ton slurry post-housing, so the loss is estimated as 0.1 kg N per ton slurry post-housing.

5.4 Alternative C1

This alternative is practically identical to Alternative P1, but here it applies for cow slurry. The separation technology used for the first separation also differs slightly. It consists of flocculation

chambers in which added polymer is mixed with the slurry; this alters the physical state of the dissolved and suspended solids and facilitates their removal by a belt press. A combination of screens and screw press is then used to finalize the separation. This technology manufactured by Kemira water (21), model Kemira 808 C for cow slurry. The flocculent used is, as in Alternative P1, cationic PAM.

Separation efficiencies were defined based on data from the technology provider except for C, Cu and Zn, for which there were no data. For C, it was assumed that the separation efficiency is the same as for DM. For Cu and Zn, the efficiencies were estimated based on Møller et al. (19) (data from screw press, with cattle slurry no.3). Since no polymer addition is involved in the study performed by Møller et al. (19), these efficiencies may be lower as those involved in the actual study, but it is yet a better approximation than simply ignoring Cu and Zn for the rest of the analysis.

Table S6 presents the separation efficiency considered for this separation technology, as well as the mass balances allowing to determine the composition of the separated liquid and solid fractions (for the first separation). Minor inconsistencies may occur in this table due to rounding. The original calculations have been performed with all the decimals. The mass balances for the second separation are presented in section 10.

Table S6. Mass balance for first separation in Alternative C1 (Kemira water technology)

	Amount in slurry before separation		Separation efficiency	<i>Mass balance: amount transferred to the solid fraction (SF)</i>	<i>Mass balance: amount transferred to the liquid fraction (LF)</i>	Solid fraction (SF) composition ^a	Liquid fraction (LF) composition ^b
Unit	kg	ton ⁻¹	%	kg ton ⁻¹ post housing	kg ton ⁻¹ post housing	kg ton ⁻¹ solid fraction	kg ton ⁻¹ liquid fraction
Total mass	1000		28.9	289	710.8	1000	1000
Dry matter (DM)	113.2		79.2	89.7	23.5	310.0	33.1
Total nitrogen (N)	6.34		50.0	3.2	3.2	11.0	4.5
Phosphorus (P)	1.03		68.6	0.7	0.3	2.4	0.5
Potassium (K)	5.90		20.0	1.2	4.7	4.1	6.6
Carbon (C)	49.7		79.2	39.4	10.3	136.1	14.5
Copper (Cu)	0.01		9.0	0.001	0.01	0.004	0.02
Zinc (Zn)	0.02		11.1	0.003	0.02	0.009	0.03

^a Calculated as: (amount transferred to the solid fraction × 1000 kg ton⁻¹) / mass amount transferred to the solid fraction. ^b Calculated as: (amount transferred to the liquid fraction × 1000 kg ton⁻¹) / mass amount transferred to the liquid fraction.

6. Process Flow Diagram for Dairy Cow Slurry Scenario

The process flow diagram for dairy slurry scenarios (reference and biogas alternative) is presented in Figure S1. In this figure, all involved flows are related to the functional unit, i.e. the excreted 1 ton of cow slurry.



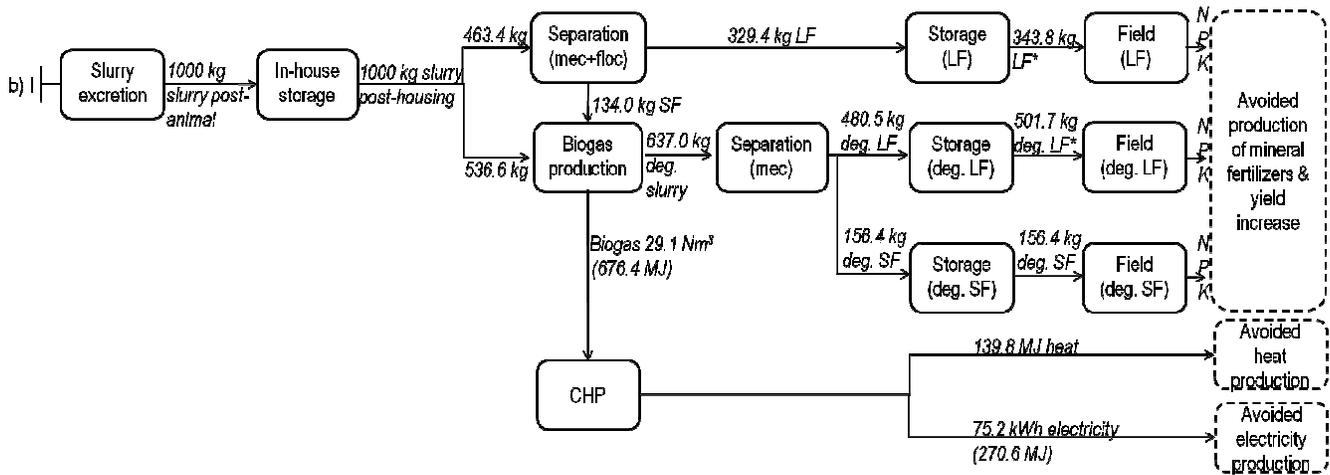


Figure S1. Process flow diagrams of the alternatives compared for dairy cow slurry management: (a) Reference system (REF-cow), (b) Alternative C1. The dotted lines indicate avoided processes. Flows marked with * include the addition of rain water. The diagrams are simplified and only include the main processes involved in the model. All flows are related to the functional unit.

7. Identification of Marginals

The two main marginals to identify in this study relate with the (avoided) mineral fertilizers and the energy (electricity and heat). Table S7 summarizes the marginal processes used in this study.

Table S7. Description of the marginal processes used in this study

Marginal process	Description	Market trend and scope considered	Remark
Electricity	Mix electricity marginal: 1% wind; 48 % coal at power plant; 51 % natural gas at power plant.	Rising trend; Denmark	This acknowledges the concept of complex marginal technologies introduced by (22). The marginal electricity was identified based on a comprehensive energy system analysis for the Danish energy system performed through the use of the EnergyPLAN model (23). The complex electricity marginal selected is adapted from the simulation performed by (24).
Heat	100 % coal	Fluctuating; local	Considering that the biogas plant is connected to the district heating grid, involving that the heat from the biogas plant replaces the marginal energy source of the CHP producing plant. Moreover, it was considered that only 60 % of the surplus from the biogas plant (i.e. after uses for the process itself) is used, in order to reflect the seasonal variations in the demand for heat in Denmark.
N fertilizer	Ammonium nitrate, as N	Rising trend; North European market	Based on medium and long term forecasts (25-26), an increase in mineral N consumption is likely, both in Europe and worldwide. Assuming that the consumption pattern from the past 10 years (27) reflects competitiveness, ammonium nitrate is identified as the marginal fertilizer.
P fertilizer	Diammonium phosphate, as P ₂ O ₅	Rising trend; World market	Based on long term forecasts (26), the trend for P consumption is rising. Based on (28), diammonium phosphate units are envisioned to represent a significant proportion of the new capacity installed, besides to be the P fertilizer with the greatest apparent consumption for the last decade (29).
K fertilizer	Potassium chloride, as K ₂ O.	Rising trend; World market	Long terms projections for K fertilizers consumption also indicate an increased trend, for EU and worldwide (26). Potassium chloride accounts for about 95 % of all K fertilizers used in agriculture, being the cheapest per ton (30).

8. Life Cycle Inventory Methodology for Emission Flows

8.1 In-house slurry storage

The methodologies used for assessing the losses in the housing units are presented in Table S8, for both pig and cow slurry systems. Substances targeted are methane (CH₄), carbon dioxide (CO₂),

ammonia (NH₃), dinitrogen monoxide (N₂O), nitrogen (N₂), nitrogen monoxide (NO) and nitrogen oxides (NO_x).

Table S8. Methodology used for inventory: in-house slurry storage.

Substance	Description of the methodologies used for each substance flows
<i>Emission to air</i>	
CH ₄	IPCC Tier 2 approach (16), considering a methane conversion factor (MCF) of 17 %. The maximum methane producing capacity (B ₀) considered are of 0.45 and 0.24 m ³ CH ₄ per kg VS excreted for pig and dairy cow slurry, respectively. The amount of VS post-animal is from Table S1.
CO ₂	Estimated as total losses of C in-house (from Table S1) minus C loss as CH ₄ .
NH ₃ -N	Based on (13), NH ₃ -N is estimated as 16 % of the total N post-animal for pig slurry and as 8 % of the total N post-animal for dairy cow slurry.
N ₂ O-N (direct)	Based on IPCC guidelines (16), 0.002 kg N ₂ O-N are emitted per kg of N in post-animal slurry. This stands for both pig and dairy cow slurry.
N ₂ O-N (indirect, from NH ₃ and NO _x)	Based on IPCC guidelines (16), 0.01 kg N ₂ O-N are emitted per kg of (NH ₃ -N + NO _x -N) volatilized. This stands for both pig and dairy cow slurry.
N ₂ -N	Estimate derived from (17), consisting of assuming that N ₂ -N = (direct) N ₂ O-N x 3. This stands for both pig and dairy cow slurry.
NO-N and NO _x -N	Estimate derived from (17), consisting of assuming that NO-N = (direct) N ₂ O-N x 1. This stands for both pig and dairy cow slurry. As NO _x = NO + NO ₂ , and as no data were available to estimate NO ₂ , it is assumed that NO-N = NO _x -N.
<i>Discharges to soil and water</i>	
Assumed negligible, based on Danish conditions.	

Based on the methodologies presented in Table S8, the life cycle inventory can be performed; this is presented in Hamelin et al. (31). As the biogas alternatives do not involve changes in the housing units, the inventory is the same for the alternatives and the reference, for pig and cow slurry, respectively.

As indicated in the manuscript, it is likely that the CH₄ emissions from in-house slurry storage have been slightly overestimated. The methodology used to estimate the emissions of CH₄ from the slurry stored in the housing units is based on IPCC guidelines (16). This methodology involves a “methane conversion factor” (MCF), which ranges between 0 % (no methane formation) to 100 % (the full

methane producing potential is achieved). The present study used a rather conservative MCF (17 %), the alternative being a MCF of 3 %. This lower MCF would imply 82 % lower CH₄ losses from in-house storage as well as increased subsequent production of biogas from higher slurry C content. Although these figures are significant, the choice of the MCF has no influence on the overall conclusions as the in-house slurry storage process is equal in all scenarios. It is nevertheless acknowledged that other approaches, like using an Arrhenius relationship as proposed by (1, 32), may have been used instead of the IPCC methodology.

8.2 Outdoor Storage

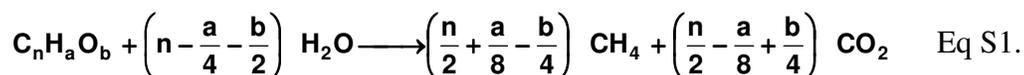
Table S9 presents the methodologies used for assessing the losses during outdoor storage, for all the different slurry fractions involved. For pigs, it is assumed that the raw slurry, the LF, the degassed liquid fraction (deg. LF) as well as the degassed slurry (deg. slurry) are stored in a leakage free concrete slurry tank covered by a floating layer of straw (2.5 kg of straw per ton slurry stored). This assumption was also made for cow LF and deg. LF, while for raw cow slurry no straw is added as it is assumed that the natural crust cover forming by itself is a sufficient cover under Danish conditions. For both cow and pig degassed solid fraction (deg. SF), it is assumed the deg. SF is stored as a heap lying on a concrete slab, covered by a plastic sheet in order to reduce the degradation of organic matter favoured when the heap is exposed to air (33-35). Emissions from storage of ashes in Alternative P3 are considered insignificant, as well as emissions from temporal storage of raw slurry, SF and FP prior their use as an input for biogas production.

Table S9. Methodology used for inventory: outdoor slurry storage

Substance	Description of the methodology used for each fraction types			
	Raw slurry	LF	Deg. slurry and deg. LF	Deg. SF
Emission to air				
CH ₄	As in Table S8, but with a MCF of 10%.	Same methodology as for raw slurry storage (MCF of 10 %), but with the VS content of the LF.	Same methodology as for raw slurry (MCF of 10 %), but with the VS content of the deg. slurry. Also, a reduction potential factor of 50 % is applied, based on (36) in order to account for the fact that the remaining VS are mostly slowly degradable VS.	CH ₄ -C is estimated as 0.17 % of the C in the deg. SF to store, based on (33)
N ₂ O-N (direct)	Emission of 0.005 kg N ₂ O-N per kg N in slurry post-animal, based on IPCC guidelines (16).	Rough estimate based on emissions from raw slurry, adjusted with relative N ratios of LF and raw slurry.	Same methodology as for LF, but a reduction factor of 40 % is applied, based on (36), to account for the effect of digestion.	Estimated as 0.04 % of the total N in the deg. SF to store, based on (33).
CO ₂	Estimated as total losses of C during storage (from Table S1) minus C loss as CH ₄ .	Calculated from CH ₄ emissions, based on the Buswell equation (37) and the distribution of the organic components constituting the VS in slurry, see Tables S10-S11. Pig: 1.42 kg CO ₂ per kg CH ₄ ; Cow: 1.67 kg CO ₂ per kg CH ₄ .		CO ₂ -C estimated as 1.9 % of the C in the deg. SF to store, based on (33).
NH ₃ -N	Based on (13), emissions of NH ₃ -N are 2 % of the total N in the slurry input for storage.			Pig: estimated as 13 % of the total N in the deg. SF to store, based on (38). Cow: estimated as 5.75 % of the total N in the deg. SF to store, based on an average from recent studies (34-35).
N ₂ O-N (indirect, from NH ₃ and NO _x)	Same methodology as described in Table S8.			
N ₂ -N	Same methodology as described in Table S8.			
NO-N	Same methodology as described in Table S8.			
Discharges to soil and water				
Assumed negligible, based on Danish conditions.				

^a The CO₂ from raw slurry is not calculated with this ratio in order to keep the mass balance consistent. This is because of the data used to establish the C content of the reference slurry composition, which involved a “backwards” calculation to pass from slurry post-storage to slurry post-animal.

An original methodology has been developed in order to assess the biogenic CO₂ emissions from storage. Thus, biogenic CO₂ emissions have been estimated as a function of biogenic CH₄ releases. The ratio between CO₂ and CH₄ emitted during anaerobic degradation is estimated based on the Buswell equation (37), as presented in Equation S1:



The organic components making up the VS in slurry and their relative amount in pig and cow slurry were taken from Sommer et al. (1), and are presented in Table S10.

Table S10. Organic components constituting the VS in slurry and their relative amount in pig and cow slurry (adapted from Sommer et al. (1)).

Organic component	Formula	Relative amount in pig slurry (%)	Relative amount in cow slurry (%)
<i>VS easily degradable</i>			
VS lipid	C ₅₇ H ₁₀₄ O ₆	16.2	7.7
VS protein	C ₅ H ₇ O ₂ N	27.0	16.8
VS Volatile fatty acids (VFA)	C ₂ H ₄ O ₂	8.5	4.0
VS carbohydrates easily degradable	C ₆ H ₁₀ O ₅	27.1	41.5
<i>VS slowly degradable</i>			
VS carbohydrates slowly degradable	C ₆ H ₁₀ O ₅	21.2	30.1
TOTAL		100	100.1 ^a

^a Based on values from Sommer et al. (1), the sum corresponds to 100.1 % instead of 100 %, which may be due to a rounding error. For the calculations in this study, it is assumed that the error was for the heavily degradable carbohydrates (i.e. 30.0 % instead of 30.1 %).

Based on Equation S1 and Table S10, the ratio between the number of moles of CO₂ and CH₄ from the full degradation of the easily degradable VS in the slurry can be calculated, as presented in Table S11.

Table S11. Calculation of the ratio between biogenic CH₄ and CO₂ resulting from the degradation of the easily degradable VS in the slurry.

Organic component	Unit	Pig slurry		Cow slurry	
		CH ₄	CO ₂	CH ₄	CO ₂
VS lipid	moles of CH ₄ and CO ₂ from the degradation of 1 mole VS lipid (moles)	40	17	40	17
	Relative amount in the slurry (%)	16.2	16.2	7.7	7.7
	Moles of CH ₄ and CO ₂ from the degradation of 1 mole VS lipid, as weighted for pig and cow slurry (moles)	6.48	2.75	3.08	1.31
VS protein	moles of CH ₄ and CO ₂ from the degradation of 1 mole VS protein (moles)	2.9	2.1	2.9	2.1
	Relative amount in the slurry (%)	27.0	27.0	16.8	16.8
	Moles of CH ₄ and CO ₂ from the degradation of 1 mole VS protein, as weighted for pig and cow slurry	0.78	0.57	0.48	0.36
VS VFA	moles of CH ₄ and CO ₂ from the degradation of 1 mole VS VFA	1	1	1	1
	Relative amount in the slurry	8.5	8.5	4.0	4.0
	Moles of CH ₄ and CO ₂ from the degradation of 1 mole VS VFA, as weighted for pig and cow slurry	0.09	0.09	0.04	0.04
VS carbohydrates easily degradable	moles of CH ₄ and CO ₂ from the degradation of 1 mole VS carbohydrates easily degradable	3	3	3	3
	Relative amount in the slurry	27.1	27.1	41.5	41.5
	Moles of CH ₄ and CO ₂ from the degradation of 1 mole VS carbohydrates easily degradable, as weighted for pig and cow slurry	0.81	0.81	1.25	1.25
SUM (moles of CH ₄ and CO ₂ as weighted for pig and cow slurry)		8.16	4.22	4.85	2.96
Ratio CO ₂ : CH ₄		0.52 moles CO ₂ per mole CH ₄		0.61 moles CO ₂ per mole CH ₄ .	
Amount of CO ₂ (g) per g of CH ₄		1.42 g CO ₂ per g CH ₄		1.67 g CO ₂ per g CH ₄	

Based on the methodologies presented in Table S9, the life cycle inventory can be performed for the storage of the different slurry fractions; this is presented in Hamelin et al. (31)

8.3 Field processes

The procedure used for estimating the losses related to field processes is presented in Table S12 (emissions to air) and S13 (discharges to soil and water). Emission flows related to soil C changes were calculated considering a 100 years horizon for soil C as well as a sandy soil (soil JB3 of the Danish soil classification).

Table S12. Methodology used for inventory: field processes, emissions to air.

Substance	Description of the methodology used for each fraction types				
	Raw slurry	LF	Deg. slurry	Deg. SF	Deg. LF
<i>Emission to air</i>					
CH ₄	Assumed negligible, based on field experiments results (39-40).				
N ₂ O-N (direct)	Emission of 0.01 kg N ₂ O-N per kg N in slurry post-storage, based on IPCC guidelines (41).				Emission of N ₂ O-N of 0.4 % of the applied N, based on (42).
CO ₂	Modeled by the 3-pooled dynamic soil model C-TOOL (43-44).				
NH ₃ -N	Emissions of NH ₃ -N are 0.138 kg NH ₃ -N per kg NH ₄ -N (for pigs) and 0.217 kg NH ₃ -N per kg NH ₄ -N (for cows), based on an area and slurry-N weighted average of all NH ₃ -N losses in the crop rotation defined for the pig/cow slurry scenario ^a .	Emissions of NH ₃ -N calculated as for raw slurry, but a reduction potential factor of 50 % is applied, based on (38), to account for the fact that LF has a low DM content and infiltrates faster than raw slurry.	Estimated with the same methodology as for raw pig slurry.	Emission of NH ₃ -N are 40 % of the NH ₄ -N applied ^b .	Estimated with the same methodology as for raw (pig and cow) slurry.
N ₂ O-N (indirect, from NH ₃ and NO _x)	Same methodology as described in Table S8.				
N ₂ O-N (indirect, from N leaching)	Based on IPCC guidelines (41), 0.0075 kg N ₂ O-N are emitted per kg of N leaching. This stands for both pig and dairy cow slurry.				
NO _x -N	Based on (45), emissions of NO _x -N correspond to 10 % of the direct N ₂ O-N emissions.				
N ₂ -N	Estimated from SimDen model ratios between N ₂ -N and N ₂ O-N of 3:1, for sandy soils (46).				

^a Crop rotation is as described in section 3. NH₄-N is estimated as 79 % and 58 % of total N in raw slurry to be applied, for pig and cow slurry, respectively (38). ^b Assuming the application takes place during the spring and that the applied degassed fibre fraction is ploughed or harrowed within 6 hours after the application. NH₄-N of deg. SF is assumed to be 25 % of the N content of the deg. SF to be applied, based on (38).

Table S13. Methodology used for inventory: field processes, discharges to soil and water

Substance	Description of the methodology used for each fraction types				
	Raw slurry	LF	Deg. slurry	Deg. SF	Deg. LF
<i>Discharges to soil and water</i>					
N leaching	Corresponds to: 51.2 % of [N in raw slurry to be applied minus NH ₃ -N losses] for pig and; 53.5 % of [N in raw slurry to be applied minus NH ₃ -N losses] for cow. Based on (31).	As for raw slurry, but assuming 21 % of the N affect the soil as raw slurry and 79 % of the N affect the soil as mineral N, for pig. For cow, these proportions are 42 % and 58 %, respectively. The factor for N leaching from mineral fertilizer is 46.8 % ^a .	Estimated as for raw pig slurry.	Based on calculations with C-TOOL, there is, after NH ₃ losses, 5.80 and 5.48 kg N left for harvest and leaching, for pig and cow, slurry, respectively. The proportion of this N that ends up leaching is assumed as for raw slurry (51.2 % for pig and 53.5 % for cow).	As for LF, but assuming 49 % of the N affects the soil as raw slurry and 51 % of the N affects the soil as mineral N, for pig. For cow, these proportions are 38 % and 62 %, respectively.
P leaching	P leaching to soil corresponds to 10 % of the P applied to the field, and 6 % of this P reach the aquatic recipients, based on (47).				
Cu	All Cu applied to soil is assumed to leach.				
Zn	All Zn applied to soil is assumed to leach.				

^a The marginal response in terms of N partitioning between the different N fates following field application of mineral N, pig slurry-N and cow slurry-N were established, based on calculations of soil N changes performed with C-TOOL. These estimates are for sandy soil considering a 100 years horizon for C turnover. For LF and deg. LF, the proportion affecting the soil as raw slurry is based on the C:N ratio of LF post-storage divided by the C:N ratio of the raw slurry post-storage, and the remaining is the proportion affecting the soil as mineral N. See Hamelin et al. (31) for additional details.

Based on the methodologies presented in Table S12 and S13, the life cycle inventory can be performed for the field processes related to the use of the different slurry fractions; this is presented in Hamelin et al. (31)

8.4 Energy Consumption

The energy consumption of the different processes involved in this life cycle assessment has been considered and is summarized in Table S14.

Table S14. Summary of data used for energy consumption

Life cycle stage	Unit	Specifications	Value	Comments
Slurry transfer from housing units to separation or outdoor storage	kWh ton ⁻¹ slurry post-housing		1.7	Including 1.2 kWh for stirring and 0.5 kWh for pumping
Slurry separation	kWh ton ⁻¹ slurry input in the separator or in the pellets process	Decanter centrifuge (P1 and C1)	2.18	Based on (48)
		Screw press (P2)	0.95	Data from technology supplier
		Pellets fabrication (P3)	19 ^a	Data from technology supplier. This includes the energy for separation.
Outdoor storage	kWh ton ⁻¹ stored slurry	Raw pig slurry, deg. slurry	2.9	Including 1.2 kWh for stirring when straw is added as a cover, 1.2 kWh for stirring before pumping for transfer to field and 0.5 for pumping.
		Raw cow slurry	1.7	As for raw pig slurry, but without the straw addition.
		LF, deg. LF	1.45	To account for lower DM content, it is estimated as 50 % of the consumption for raw pig slurry.
		deg. SF	0	No energy involved.
Slurry application in the field	kg diesel ton ⁻¹ material applied	LF, deg. LF, deg. slurry, raw pig and cow slurry	0.34	Based on a personal communication with Mogens Kjelddal, Landsforeningen Danske Maskinstationer, March 2009.
		deg. SF	0.53	Based on (45)
Application of mineral fertilizers	kg diesel ton ⁻¹ fertilizer applied	For mineral N, P and K	0.006	Based on (49)
Transport of slurry	kg diesel ton ⁻¹ material applied km ⁻¹	For any slurry or slurry fraction transported	0.044	Based on (45)

^a The heat needed for drying the fibres comes from the heat produced when some of the produced pellets are combusted, corresponding to 120 MJ per ton slurry post-housing.

9. Biogas Production and Energy Balance

9.1 Biogas composition, lower heating value, biogas density and description of the plant

The biogas produced is considered to be composed of 65 % CH₄ and 35 % CO₂. This composition implicitly assumes that other gases (e.g. N₂, O₂, H₂S, H₂O, CO, H₂), which altogether generally account for less than 1 % of the biogas composition, can be neglected.

Based on these proportions of CH₄ and CO₂ in the biogas and considering a CH₄ density of 0.717 kg Nm⁻³ and a CO₂ density of 1.977 kg Nm⁻³, the biogas density is calculated as 1.158 kg Nm⁻³. Similarly, based on a heat value for CH₄ of 9.94 kWh Nm⁻³, the biogas lower heat value (LHV) was calculated as 6.46 kWh Nm⁻³ (23.36 MJ Nm⁻³).

The biogas plant considered in this study consists of bioreactors for the biogas production, of receiving facilities and storage tanks for SF, raw and degassed slurry and of a co-generation unit allowing to produce heat and electricity from the biogas. A two-step digestion with an annual treatment capacity of 100 000 m³ of biomass is considered for the calculations. Both steps are continuously operated and fully mixed in overflow tanks with a hydraulic retention time defined by the ratio between the digester volume and the daily biomass input volume.

The first step yields 90 % of the final biogas yield and is a carefully controlled process in terms of temperature, retention time and loading. The second step is a covered post-digestion tank without temperature control and with a relatively low loading. The biogas plant is an air-tight system and therefore principally without any uncontrolled gaseous emissions. In this study, it is considered that the anaerobic digestion operates at mesophilic temperatures, i.e. around 37 °C. The biogas is burned in a gas engine with efficiencies of 46 % for heat and 40 % for electricity (50), for a total efficiency of 86 %.

9.2 Calculation of the share of raw slurry and solid fraction in the mixture input

For all scenarios, the amount of raw slurry and solid fraction (or fibre pellets) in the mixture input for biogas production is determined in order to obtain a biomass mixture that has a DM of 10 % after the first digestion step. This is based on personal communication with an expert operating at several biogas plants in Denmark (personal communication with Anders Peter Jensen, Xergi, June 2009). This involves

that the proportions of solid fraction (or fibre pellets) and raw slurry shall be found so the ratio DM: total weight is equal to 0.1. To solve this, a second equation is introduced, i.e. the sum of raw slurry and solid fraction added should equal 1 ton. This therefore corresponds to a system with two equations and two unknown:

$$0.1 = \frac{((W_{\text{raw}} \times \text{DM}_{\text{raw}}) - \text{VS}_{\text{deg,raw}}) + ((W_{\text{SF}} \times \text{DM}_{\text{SF}}) - \text{VS}_{\text{deg,SF}})}{(W_{\text{raw}} - W_{\text{biogas,raw}}) + (W_{\text{SF}} - W_{\text{biogas,SF}})} \quad \text{Eq S2.}$$

$$1000 = W_{\text{raw}} + W_{\text{SF}} \quad \text{Eq S3.}$$

Where W_{raw} is the weight of the raw slurry input (kg); DM raw is the dry matter content of the raw slurry (%); $\text{VS}_{\text{deg,raw}}$ are the VS degraded from the raw slurry (kg); W_{SF} is the weight of the solid fraction or fibre pellets (kg); DM_{SF} is the dry matter content of the solid fraction or fibre pellets (%); $\text{VS}_{\text{deg,SF}}$ are the VS degraded from the solid fraction or fibre pellets (kg); $W_{\text{biogas,raw}}$ is the weight of biogas (kg).

The degraded VS for each fraction types (raw slurry or slurry fraction) can be calculated based on the assumption that the VS represents 80 % of DM (51), and using the degradation rate (DR) (in % of the VS) that applies for the slurry or slurry fraction under consideration. This is however only for the first digestion step (and not for the total produced), as the calculations are based for the first digestion step. This is presented in Equation S4.

$$\text{VS}_{\text{deg,fract}} = (W_{\text{fract}} \times \text{DM}_{\text{fract}}) \times 80\% \times \text{DR}_{\text{fract}} \quad \text{Eq S4}$$

Where $\text{VS}_{\text{deg,fract}}$ is the amount of VS degraded for a given fraction during the first digestion step (kg), W_{fract} is the weight of the fraction, DM_{fract} is the DM content of the fraction (%) and DR_{fract} is the degradation rate of the fraction (in % of the VS) for the first digestion step. Degradation rates and calculation of VS degraded for each fractions are presented in Table S15 (for the first digestion step).

Table S15. Degradation rates and calculation of the VS degraded for each fraction, for the first digestion step

	DM _{fract} (%)	Total VS input (kg)	DR _{fract} (% of the VS)	VS _{deg,fract}
Raw pig slurry	7.0	$W_{\text{raw}} \times 0.07 \times 0.8 = 0.056 \times W_{\text{raw}}$	60	$0.03 \times W_{\text{raw}}$
Raw cow slurry	11.3	$W_{\text{raw}} \times 0.113 \times 0.8 = 0.09 \times W_{\text{raw}}$	46.7	$0.04 \times W_{\text{raw}}$
SF, Alternative P1	26.6	$W_{\text{SF}} \times 0.266 \times 0.8 = 0.213 \times W_{\text{SF}}$	60	$0.13 \times W_{\text{SF}}$
SF, Alternative C1	31.0	$W_{\text{SF}} \times 0.31 \times 0.8 = 0.248 \times W_{\text{SF}}$	46.7	$0.11 \times W_{\text{SF}}$
SF, Alternative P2	39.7	$W_{\text{SF}} \times 0.397 \times 0.8 = 0.318 \times W_{\text{SF}}$	37.8	$0.12 \times W_{\text{SF}}$
FP, Alternative P3	88.9	$W_{\text{SF}} \times 0.889 \times 0.8 = 0.711 \times W_{\text{SF}}$	37.8	$0.27 \times W_{\text{SF}}$

The weight of the biogas after the first digestion step can be determined based on the methane yield (for the first digestion step only), the total VS input, the biogas density (i.e. 1.158 kg Nm⁻³) and the volumetric content of CH₄ in the biogas (0.65 Nm³ CH₄ Nm⁻³ biogas, based on the biogas composition).

Methane yields for the first digestion step are presented in Table S16, as well as the calculations for determining the weight of the biogas for all slurries and slurry fractions.

Table S16. Methane yields for the first digestion step and calculation of the biogas weight for all slurries and slurry fractions

		Total input (kg)	VS	CH ₄ yield for the first digestion step (Nm ³ CH ₄ ton ⁻¹ VS)	Calculation	Biogas weight (kg)
Raw slurry	pig	$0.056 \times W_{\text{raw}}$	290	290	$(0.056 \times W_{\text{raw}} \times 290 \times 1.158) / (0.65 \times 1000)$	$0.0289 \times W_{\text{raw}}$
Raw slurry	cow	$0.09 \times W_{\text{raw}}$	210	210	$(0.09 \times W_{\text{raw}} \times 210 \times 1.158) / (0.65 \times 1000)$	$0.0337 \times W_{\text{raw}}$
SF, Alternative P1		$0.213 \times W_{\text{SF}}$	290	290	$(0.213 \times W_{\text{SF}} \times 290 \times 1.158) / (0.65 \times 1000)$	$0.110 \times W_{\text{SF}}$
SF, Alternative C1		$0.248 \times W_{\text{SF}}$	210	210	$(0.248 \times W_{\text{SF}} \times 210 \times 1.158) / (0.65 \times 1000)$	$0.093 \times W_{\text{SF}}$
SF, Alternative P2		$0.318 \times W_{\text{SF}}$	170	170	$(0.318 \times W_{\text{SF}} \times 170 \times 1.158) / (0.65 \times 1000)$	$0.096 \times W_{\text{SF}}$
SF, Alternative P3		$0.711 \times W_{\text{SF}}$	170	170	$(0.711 \times W_{\text{SF}} \times 170 \times 1.158) / (0.65 \times 1000)$	$0.215 \times W_{\text{SF}}$

Using Equation S3 and expressing W_{raw} as $1000 - W_{\text{SF}}$, Equation S2 can be solved. The results are presented in Table S17. It should be noted that the values presented in Table S17 have been calculated without cutting any decimals. Because of this, minor inconsistencies may occur if calculations are made with the rounded values presented in Tables S15 and S16.

Table S17. Input of raw slurry and solid fraction in the digester for all biogas scenarios

	W_{raw} (kg)	W_{SF} (kg)	Share of the raw slurry in the input (%)	Share of the solid fraction in the input (%)
Alternative P1	445	555	44.5	55.5
Alternative P2	753	247	75.3	24.7
Alternative P3	899	101	89.9	10.1
Alternative C1	800	200	80.0	20.0

9.3 Calculation of the total biogas produced

Based on the values for W_{raw} and W_{SF} , as well as on the overall CH_4 yields (for both degradation steps), the volumetric proportion of CH_4 in the biogas, the DM content of all fractions and the assumption that VS are 80 % of the DM, the total amount of biogas produced can be calculated. This calculation, together with the overall CH_4 yields, is presented in Table S18.

Table S18. Calculation of the total volume of biogas produced for all biogas alternatives

	Alternative P1	Alternative P2	Alternative P3	Alternative C1
<i>Raw slurry</i>				
Input (kg)	445	753	899	800
DM (kg ton ⁻¹ slurry input)	69.7	69.7	69.7	113.2
CH ₄ yield (Nm ³ CH ₄ ton ⁻¹ VS)	319	319	319	231
Calculation	$(445 \times 69.7 \times 0.80 \times 319) / (0.65 \times 1000 \times 1000)$	$(753 \times 69.7 \times 0.80 \times 319) / (0.65 \times 1000 \times 1000)$	$(899 \times 69.7 \times 0.80 \times 319) / (0.65 \times 1000 \times 1000)$	$(800 \times 113.2 \times 0.80 \times 231) / (0.65 \times 1000 \times 1000)$
Biogas produced (Nm ³)	12.2	20.6	24.6	25.7
<i>Solid fraction</i>				
Input (kg)	555	247	101	200
DM (kg ton ⁻¹ slurry input)	265.9	396.9	889.3	310.0
CH ₄ yield (Nm ³ CH ₄ ton ⁻¹ VS)	319	187	187	231
Calculation	$(555 \times 265.9 \times 0.80 \times 319) / (0.65 \times 1000 \times 1000)$	$(247 \times 396.9 \times 0.80 \times 187) / (0.65 \times 1000 \times 1000)$	$(101 \times 889.3 \times 0.80 \times 187) / (0.65 \times 1000 \times 1000)$	$(200 \times 310.0 \times 0.80 \times 231) / (0.65 \times 1000 \times 1000)$
Biogas produced (Nm ³)	57.9	22.5	20.7	17.6
Total biogas produced (Nm ³ ton ⁻¹ input mixture)	70.1	43.1	45.3	43.3

9.4 Energy balance

Based on the total biogas produced, on the heating value of the biogas (6.46 kWh Nm⁻³ or 23.26 MJ Nm⁻³) and on the efficiency of the engine for heat and electricity (40 % for electricity, 46 % for heat), the gross energy produced from the biogas produced can be calculated (i.e. before a share of the produced heat is used for the process itself). This is presented in Table S19.

Table S19. Gross energy produced from the biogas

	Total biogas produced (Nm ³ ton ⁻¹ input mixture)	Electricity produced (kWh ton ⁻¹ input mixture)	Gross heat (MJ ton ⁻¹ input mixture)
Alternative P1	70.1	181.1	749.9
Alternative P2	43.1	111.4	461.1
Alternative P3	45.3	117.1	484.6
Alternative C1	43.3	111.9	463.2

An electricity input is needed for producing the biogas, i.e. for pumping, stirring, etc. In this study, the electricity input for producing the biogas is estimated as 5 % of the net energy production. This is based on measurements performed at several Danish biogas plants (personal communication with Anders Peter Jensen, Xergi, June 2009). Based on this, the internal electricity consumption can be calculated, as presented in Table S20.

Table S20. Internal electricity consumption for all biogas alternatives

	Total biogas produced (Nm ³ ton ⁻¹ input mixture)	Internal electricity consumption (kWh ton ⁻¹ input mixture)
Alternative P1	70.1	9.06
Alternative P2	43.1	5.57
Alternative P3	45.3	5.85
Alternative C1	43.3	5.59

The heat consumption was calculated assuming the mixture should be heated from 8°C (average temperature in Denmark) and 37°C (process temperature), which represents a difference of 29°C. As the plant considered is well insulated, no heat losses are assumed. Based on a specific heat for the DM of 3.00 kJ kg⁻¹°C⁻¹ and of 4.20 kJ kg⁻¹°C⁻¹ for water as well as on the DM and water content of the mixture input to the biogas plant, the heat consumption can be calculated. The DM and water content of the mixture input can be calculated based on the proportion of each fraction put into the biogas (Table S17) and on the DM content of these (Table S1, Tables S3-S6). This is presented in Table S21, together with the calculation of the heat consumed to run the biogas process.

Table S21. Internal heat consumption for all biogas alternatives

	DM from raw slurry (kg mixture input) (kg ton ⁻¹ mixture input)	DM from solid fraction (kg mixture input) (kg ton ⁻¹ mixture input)	Total DM (kg mixture input) (kg ton ⁻¹ mixture input)	Total water ^a (kg mixture input) (kg ton ⁻¹ mixture input)	Total heat consumption (MJ mixture input) (MJ ton ⁻¹ mixture input)	Ratio heat consumed: heat produced (%)
Alternative P1	(445×69.7)/1000 = 31	(555×265.9)/1000 = 148	179	821	115.6	15
Alternative P2	(753×69.7)/1000 = 52	(247×396.9)/1000 = 98	150	850	116.5	25
Alternative P3	(899×69.7)/1000 = 63	(101×889.3)/1000 = 90	153	847	116.5	24
Alternative C1	(800×113.2)/1000 = 91	(200×310.0)/1000 = 62	153	847	116.5	25

^a Estimated as all the non-DM share of the 1 ton mixture input.

Of all the heat surpluses produced, i.e. the difference between the heat produced and the heat consumed for the process, it is considered that 60 % of it will replace marginal heat on the national grid, while the remaining 40 % corresponds to heat waste, for which there is simply no demand (e.g. during the summer). Table S22 summarizes the energy balance.

Table S22. Energy balance summary

	Electricity		Heat				
	(kWh ton ⁻¹ input mixture)		(MJ ton ⁻¹ input mixture)				
	Net electricity produced	Electricity consumed (from grid)	Gross heat produced	Heat used for the process	Net surplus heat	Net heat replacing marginal heat	Heat wasted
Alternative P1	181.1	9.06	749.9	115.6	634.3	380.6	253.7
Alternative P2	111.4	5.57	461.1	116.5	344.6	207.8	137.8
Alternative P3	117.1	5.85	484.6	116.5	368.1	220.9	147.2
Alternative C1	111.9	5.59	463.2	116.5	346.7	208.0	138.7

The values presented in Table 22 can be related to the functional unit through the flows presented in Figures 1 (manuscript) and S1.

10. Separation post biogas (alternatives P1 and C1)

Alternatives P1 and C1 involve a separation of the digested slurry post anaerobic digestion. The separation technology considered for this is the exact same as the separation technology used for the first separation in Alternative P1 (decanter centrifuge), but without the use of PAM.

Table S23 and S24 present the mass balances used to calculate the composition of the slurry after the biogas production, for Alternative P1 and C1, respectively. All nutrients and DM for the slurry entering the digester are calculated as in Table S21.

Table S23. Mass balance determining the degassed slurry composition in Alternative P1

	Composition of mixture input to the digester	<i>Mass balance: changes during biogas production</i>	<i>Mass balance: amount after biogas production</i>	Composition of degassed slurry after biogas production
Unit	kg ton ⁻¹ mixture input	kg	kg	kg ton ⁻¹ degassed slurry
Total mass	1000	-81.2 ^a	918.8	1000
Dry matter (DM)	178.6	-81.2 ^b	97.4	106.0
Total nitrogen (N)	8.0	<i>No change</i>	8.0	8.7
Phosphorus (P)	2.9	<i>No change</i>	2.9	3.2
Potassium (K)	2.3	<i>No change</i>	2.3	2.5
Carbon (C)	85.4	-38.1 ^c	47.3	51.4
Copper (Cu)	0.04	<i>No change</i>	0.04	0.04
Zinc (Zn)	0.2	<i>No change</i>	0.2	0.16

^a This loss corresponds to the biogas produced. It is expressed in mass terms through the biogas density, i.e. 1.158 kg Nm⁻³. ^b No water loss assumed therefore the change in DM is the same as the change in total mass. ^c Calculated as losses (C-CH₄ and C-CO₂) from the biogas plus the losses from the digestion process. In this study, CH₄ losses from the digestion process are calculated as 1 % of the produced methane. Biogenic CO₂ losses are calculated based the biogenic methane losses (Table S11), i.e. 1.42 kg CO₂ are emitted per kg of CH₄.

Table S24. Mass balance determining the degassed slurry composition in Alternative C1

Unit	Composition of mixture input to the digester	Mass balance: changes during biogas production	Mass balance: amount after biogas production	Composition of degassed slurry after biogas production
	kg ton ⁻¹ mixture input	kg	kg	kg ton ⁻¹ degassed slurry
Total mass	1000	-50.2 ^a	949.8	1000
Dry matter (DM)	152.6	-50.2 ^b	102.4	107.9
Total nitrogen (N)	7.3	No change	7.3	7.7
Phosphorus (P)	1.3	No change	1.3	1.4
Potassium (K)	5.5	No change	5.5	5.8
Carbon (C)	67	-23.6 ^c	43.4	45.7
Copper (Cu)	0.01	No change	0.01	0.01
Zinc (Zn)	0.02	No change	0.02	0.02

^a This loss corresponds to the biogas produced. It is expressed in mass terms through the biogas density, i.e. 1.158 kg Nm⁻³. ^b No water loss assumed therefore the change in DM is the same as the change in total mass. ^c Calculated as losses (C-CH₄ and C-CO₂) from the biogas plus the losses from the digestion process. In this study, CH₄ losses from the digestion process are calculated as 1 % of the produced methane. Biogenic CO₂ losses are calculated based the biogenic methane losses (Table S11), i.e. 1.67 kg CO₂ are emitted per kg of CH₄.

Based on the composition of the degassed pig (Alternative P1) and cow (Alternative C1) slurries, as well as on the separation efficiencies, the composition of the solid and liquid degassed fractions can be calculated. Tables S25 and S26 present the separation efficiencies as well as the mass balances allowing to determine the composition of the separated liquid and solid fractions for this second separation, for Alternative P1 and C1, respectively. Separation efficiencies are based on (48), apart from Cu and Zn, for which they were no data. These were thus based on (19). Minor inconsistencies may occur in these tables due to rounding. The original calculations have been performed with all the decimals.

Table S25. Mass balance for the second separation in Alternative P1 (decanter centrifuge without PAM)

	Amount in degassed slurry before separation	Separation efficiency	<i>Mass balance: amount transferred to the degassed solid fraction (deg.SF)</i>	<i>Mass balance: amount transferred to the degassed liquid fraction (deg.LF)</i>	Degassed solid fraction (deg.SF) composition ^a	Degassed liquid fraction (deg.LF) composition ^b
Unit	kg ton ⁻¹ degassed slurry	%	kg ton ⁻¹ post housing	kg ton ⁻¹ post housing	kg ton ⁻¹ degassed solid fraction	kg ton ⁻¹ degassed liquid fraction
Total mass	1000	24.2	242	758	1000	1000
Dry matter (DM)	106.0	60.9	64.5	41.4	267.1	54.6
Total nitrogen (N)	8.7	21.2	1.8	6.9	7.7	9.1
Phosphorus (P)	3.2	66.2	2.1	1.1	8.9	1.4
Potassium (K)	2.5	9.7	0.2	2.2	1.0	2.9
Carbon (C)	51.4	60.9	31.3	20.1	129.6	26.5
Copper (Cu)	0.04	36.2	0.02	0.03	0.065	0.036
Zinc (Zn)	0.16	42.2	0.07	0.10	0.29	0.13

^a Calculated as: (amount transferred to the degassed solid fraction × 1000 kg ton⁻¹) / mass amount transferred to the degassed solid fraction. ^b Calculated as: (amount transferred to the degassed liquid fraction × 1000 kg ton⁻¹) / mass amount transferred to the degassed liquid fraction.

Table S26. Mass balance for the second separation in Alternative C1 (decanter centrifuge without PAM)

	Amount in degassed slurry before separation	Separation efficiency	<i>Mass balance: amount transferred to the degassed solid fraction (deg.SF)</i>	<i>Mass balance: amount transferred to the degassed liquid fraction (deg.LF)</i>	Degassed solid fraction (deg.SF) composition ^a	Degassed liquid fraction (deg.LF) composition ^b
Unit	kg ton ⁻¹ degassed slurry	%	kg ton ⁻¹ post housing	kg ton ⁻¹ post housing	kg ton ⁻¹ degassed solid fraction	kg ton ⁻¹ degassed liquid fraction
Total mass	1000	24.2	242	758	1000	1000
Dry matter (DM)	107.9	60.9	65.6	42.1	267.1	55.8
Total nitrogen (N)	7.7	21.2	1.6	6.0	6.6	8.0
Phosphorus (P)	1.4	66.2	0.9	0.5	3.7	0.6
Potassium (K)	5.8	9.7	0.6	5.3	2.3	7.0
Carbon (C)	45.7	60.9	27.8	17.9	113.3	23.7
Copper (Cu)	0.01	6.7	0.0007	0.01	0.003	0.01
Zinc (Zn)	0.02	25.3	0.006	0.02	0.02	0.02

^a Calculated as: (amount transferred to the degassed solid fraction × 1000 kg ton⁻¹) / mass amount transferred to the degassed solid fraction. ^b Calculated as: (amount transferred to the degassed liquid fraction × 1000 kg ton⁻¹) / mass amount transferred to the degassed liquid fraction.

11. Avoided Production of Mineral Fertilizers

11.1 Nitrogen

The avoided amount of mineral N is based on the substitution values fixed by the Danish regulation (52). Under this, specific replacement values are considered. These are presented in Table S27.

Table S27. Substitution values for nitrogen under the Danish regulation

Slurry type	Substitution value
Raw (pig)	75 % (100 kg slurry-N replaces 75 kg mineral N)
Raw (cow)	70 % (100 kg slurry-N replaces 70 kg mineral N)
LF, portion corresponding to the amount of FP burnt	85 % (100 kg slurry-N replaces 85 kg mineral N)

As described in the manuscript, these values are not applied to the actual N content of the slurry (e.g. as assessed by measurements), but to the post-storage N values from the Danish normative system for assessing slurry composition (14), as this is what farmers do in practice. In 2008 when the calculations for this project were performed, this was 5.00 kg N ton⁻¹ slurry post-storage for pig and 6.02 kg N ton⁻¹ slurry post-storage for cow.

For the reference slurries (pig and cow), the calculation of the avoided N is rather straight forward, as presented in Table S28.

Table S28. Calculations of avoided mineral N for the reference slurries

Slurry	Avoided mineral N calculation	Unit conversion to express the avoided N per functional unit	Avoided mineral N per functional unit
Pig	5.00 kg N ton ⁻¹ slurry post-storage × 75 % = 3.75 kg N ton ⁻¹ slurry post-storage	1086 kg slurry post-storage ton ⁻¹ slurry post-animal (Figure 1 of the manuscript)	4.07 kg N ton ⁻¹ slurry post-animal
Cow	6.02 kg N ton ⁻¹ slurry post-storage × 70 % = 4.21 kg N ton ⁻¹ slurry post-storage	1044 kg slurry post-storage ton ⁻¹ slurry post-animal (Figure S1)	4.40 kg N ton ⁻¹ slurry post-animal

These values represent the amount of mineral N that the farmer would have been allowed to apply without having the slurry. For alternatives P1, P2, and C1 the avoided mineral N is the same as for the corresponding reference slurry. This is so, because the nutrients from the normative value used by the farmers (i.e. 5.00 and 6.02 kg N ton⁻¹ slurry post-storage, for pig and cow slurry, respectively) are conserved; they are simply distributed among the different slurry fractions. The demonstration for this is available in Hamelin et al. (31).

For alternative P3, the calculation is slightly different because a part of the fibre pellets produced is combusted (i.e 40 % of the pellets produced). Based on the Danish regulation, it is 85 kg mineral N that are replaced per 100 kg slurry N for the liquid fraction associated to the part burned. This results in an amount of 4.09 kg mineral N replaced per ton of slurry post-animal. The detailed calculation for this is performed in Hamelin et al. (31).

11.2 Phosphorus and Potassium

As explained in the manuscript, the N use per area is limited by the Danish regulations, but not the P and K use. This involves that a potential consequence of applying slurry up to the N limits may be that an excess of P and K is applied.

Based on the Danish regulation (applying in 2008), the limit for N to be applied is 1.4 livestock unit per ha for pig farms and 1.7 livestock units per ha for cattle. There is 0.85 dairy cow per livestock unit (heavy race) and 35 fattening pigs per livestock unit (53). Based on the Danish normative system for assessing slurry composition (13), there is 0.52 tonnes slurry per pig (post-storage) and 21.3 tonnes slurry per dairy cow (post-storage).

Based on these values as well as on the slurry composition (Table S1), the amount of slurry applied to 1 ha is 25.48 ton for pig slurry and 30.78 ton for cow slurry. The P and K applied can therefore be calculated, as presented in Table S29.

Table S29. Calculations of the P and K applied with the slurry

Slurry	Nutrient	Calculation
Pig	P	$1.04 \text{ kg P per ton slurry} \times 25.48 \text{ ton slurry ha}^{-1} = 26.50 \text{ kg P per ha}$
	K	$2.60 \text{ kg K per ton slurry} \times 25.48 \text{ ton slurry ha}^{-1} = 66.25 \text{ kg K per ha}$
Cow	P	$0.98 \text{ kg P per ton slurry} \times 30.78 \text{ ton slurry ha}^{-1} = 30.16 \text{ kg P per ha}$
	K	$5.65 \text{ kg K per ton slurry} \times 30.78 \text{ ton slurry ha}^{-1} = 173.91 \text{ kg K per ha}$

The reference crop rotation is presented in section 3 for both a pig and a cow farm, with an indication of the applied N. Table S30 presents the P and K requirements for these rotations, based on the national guidelines for fertilization (4).

Table S30. Requirements in P and K for the reference crop rotations

Rotation	Crops	P (kg ha ⁻¹)	K (kg ha ⁻¹)
Pig farm rotation (6 years rotation)	Winter barley	18	54
	Winter rape	27	90
	Winter wheat	20	70
	Wheat	20	70
	Spring barley with catch crop	22	50
	Spring barley	22	50
<i>Annual average (kg ha⁻¹)</i>		<i>21.5</i>	<i>64</i>
Cow farm rotation (5 years rotation)	Whole crop silage	25	135
	Grass clover mixture	29	210
	Grass clover mixture	29	210
	Spring barley with catch crop	22	50
	Spring barley	22	50
<i>Annual average (kg ha⁻¹)</i>		<i>25.4</i>	<i>131.0</i>

The ratio between the crop requirements and the applied amount of nutrients with the slurry can be calculated, for P and K. This is presented in Table S31.

Table S31. Calculation of the ratio between crop requirement and amount of P and K applied with slurry

Slurry	Average crop requirements for the reference rotation (kg ha ⁻¹)		Amount of nutrients applied with slurry (kg ha ⁻¹)		Ratio crop requirement: applied amount (%)	
	P	K	P	K	P	K
Pig	21.5	64	26.5	66.26	81	97
Cow	25.4	131.0	30.16	173.91	84	75

Based on Table S31, only 81 % of the P applied with pig slurry and 97 % of the K do contribute to avoid mineral P and K fertilizers, respectively, to be produced. Similarly, only 84 % of the P applied with cow slurry and 75 % of the K do contribute to avoid mineral P and K fertilizers, respectively, to be produced.

12. Cow slurry results

The impact assessment results for alternative C1, as compared to the reference cow slurry scenario (REF-cow), are presented in Figure S2. The tendencies obtained are as for alternative P1, and will therefore not be further commented.

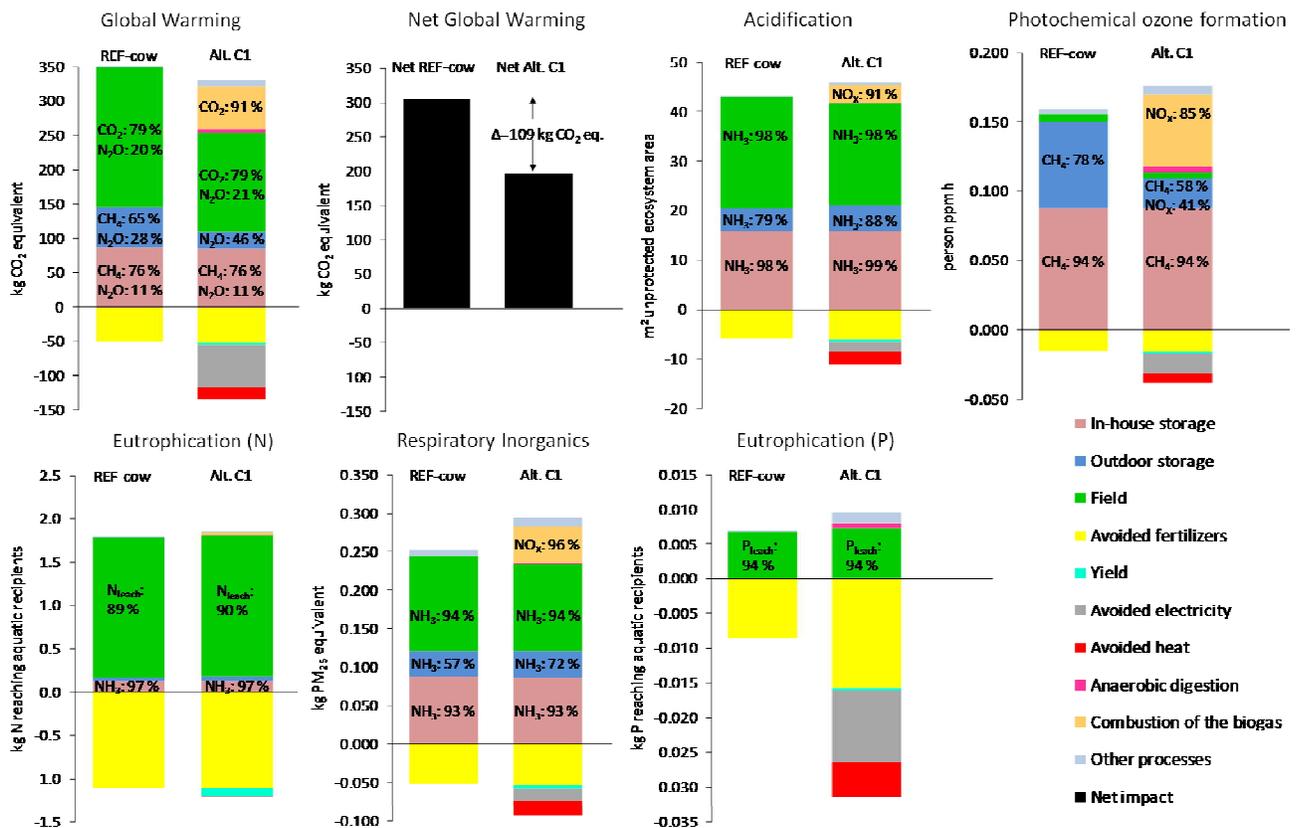


Figure S2. Breakdown of impact assessment results for all impacts, for alternative C1

The soil carbon balance for alternative C1 is presented in Table S32. For alternative C1, it is 15 % less C that ends up in the soil C pool, which is also similar to what was observed with alternative P1.

Table S32. Balance for carbon stored in the soil for alternative C1 and its reference

	REF-cow	C1
C added with slurry (kg ton ⁻¹ slurry post-animal)	47.19	33.08
C lost as CO ₂ (field) (kg ton ⁻¹ slurry post-animal)	-45.21	-31.40
C stored in the soil (kg ton ⁻¹ slurry post-animal)	1.98	1.68
Net CO ₂ -C “stored” ^a (kg ton ⁻¹ slurry post-animal)	7.26	6.16

^a This is the C stored in the soil, expressed in CO₂ through the molecular weight ratios. It does not represent a sequestration of CO₂ (it is C that is sequestered).

13. Cationic Polyacrylamide Polymer (PAM)

Polyacrylamide polymers (PAM) are widely investigated in the scientific literature as regarding their performance in solid-liquid separation of slurries (e.g. 54-59). Though the polyacrylamide polymer can be defined as many units of the monomer acrylamide, the chemical nature of the polymer and the monomer is highly different (60). While polyacrylamide is considered as a relatively safe material, the toxicity of acrylamide monomer is a major concern (61), this component being known to affect the central and peripheral nervous system (62). PAM can be charged positively (anionic), negatively (cationic) or non-charged (non-ionic) (63).

Once the PAM degrades to acrylamide monomer, the monomer is then subjected to rapid degradation in which it is decomposed to ammonia and to acrylic acid (CH₂CHCOOH), which in turn is degraded to CO₂ and water (62). Because of the extremely rapid degradation of the acrylamide monomer, it is reported that it is unlikely to find this toxic product in the environment as a result of PAM degradation (64).

Campos et al. (55) investigated if PAM degradation takes place during the anaerobic digestion of solid fractions obtained from pig slurry separated with and without the use of PAM. The authors concluded from the results of their biodegradability study that PAM is not significantly biodegradable by anaerobic microorganisms and is not toxic for anaerobic microorganisms, as no significant differences were

observed between the maximum methanogenic activity of the different treatments investigated (different concentration of PAM in the solid fractions). Similarly, Martinez-Almela and Barrera (54) as well as Gonzalez-Fernández et al. (58) also concluded that PAM residues do not contribute to toxicity of the anaerobic digestion and do not affect the methane production. Recalcitrance of PAM to microbial degradation under both aerobic and anaerobic conditions was also observed by El-Mamouni et al. (61).

Kay-Shoemake et al. (65) investigated the effect of PAM applied to agricultural soils on soil bacterial communities and nutrient cycling. They found, among others, that the bacterial numbers on soils with and without PAM application were not significantly different. They also found that PAM-treated soils planted to potatoes contained significantly higher concentrations of NO_3^- and NH_3 as compared to untreated soils. For NO_3^- , they found 36.7 mg kg^{-1} for PAM-soil as compared to 10.7 mg kg^{-1} for control soil. For NH_3 , they found 1.30 mg kg^{-1} for PAM-soil as compared to 0.50 mg kg^{-1} for control soil. This suggests that some biological degradation may take place. In an extensive review on polyacrylamide (PAM) degradation (more than 150 articles were reviewed), Caulfield et al. (60) also acknowledged this possibility (which they explained as the hydrolysis of the amide group), but they demonstrate that this degradation has to be rather limited, due to the high molecular weight of PAM that cannot pass through the biological membranes of the bacterium. This is in line with (61) who suggest that PAM may simply accumulate and persist in the environment. In their review, Caulfield et al. (60) also concluded that no evidence is existing to suggest that PAM may form free acrylamide monomer units (which are highly toxic) under biodegradation processes.

If PAM appears to be rather recalcitrant to biological degradation, it is more susceptible to undergo thermal degradation (temperatures above $200 \text{ }^\circ\text{C}$), photodegradation, chemical degradation (under very acidic or very basic conditions) as well as mechanical degradation (if submitted to high shear). These degradation processes are extensively documented in (60). In the case of application to field, photodegradation may be the most likely degradation mechanism to occur. El-Mamouni et al. (61) actually studied the degradation of PAM submitted to UV photolysis as a pre-treatment to anaerobic and biological processes. Their results indicate that this UV irradiation pre-treatment did contribute to

increase the biological degradation of PAM, under both aerobic and anaerobic conditions. However, El-Mamouni et al. (61) highlight that the irradiation conditions used in their experiment are unlikely to occur in natural environment, as they used light intensity as low as 254 nm (the lower the wavelength, the higher the energy; visible wavelength are between 400 to 700 nm) and exposition duration ranging between 12 to 72 consecutive hours.

Based on these findings, it was considered reasonable to assume, in the framework of this study, that no degradation of the PAM occur after the application of degassed PAM containing slurry fractions to the field. As linear PAM is water-soluble (64, 66), it may dissolve in water during precipitation events and leak through the water compartment. Sojka et al. (64) in fact report that very few studies have assessed the fate of PAM, as PAM cannot be easily extracted for analysis once it has been adsorbed on solid surfaces.

Due to this lack of knowledge, this study could therefore not reflect the eventual toxicity potential of the PAM accumulating in the soil. However, due to the potential toxicity impacts of PAM and concerns express relative to it (63, 67), it is suggested, for the large scale implementation of biogas from separated slurry, to favour high efficiency technologies for separating the C and VS in the solid fraction that do not involve substances with potential toxicity hazards.

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Appendix D. Supporting Information for Paper V

Supporting Information for:
Environmental Consequences of Different carbon Alternatives for Increased
Manure-Based Biogas

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83 pages, 13 figures, 76 tables

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1. Reference manure management

The reference pig slurry management considered in this study is based on the reference system described in Hamelin et al. [1], and assumed the same pre-conditions, with a few exceptions. In the present study, a partly slatted floor is considered (25-49% slatted), instead of a fully slatted floor as in [1]. The reason for this is that it is envisioned, with animal welfare regulations getting stricter [2], that more system on partly slatted floors will emerge. (In 2009, there were, in Denmark 54% of the fattening pig systems on fully slatted floors and 35% on partly slatted floor, [3]). As in [1], outdoor storage is assumed to take place in a concrete tank, covered by a straw floating layer and slurry is applied to fields with a trail-hose slurry tanker. The same reference crop rotation is considered, but updated for the amount of N required by the crop rotation (see section 6).

The reference slurry composition is based on the Danish manure standards [4], and is slightly different as the one in [1] because values have been updated for 2011. Table S1 presents the slurry composition considered, and Table S2, the life cycle inventory data considered for reference slurry management (emission flows). State-of-the-art mass balances were performed to ensure consistency between the slurry composition, and the emission flows. Based on Table S1, there is 1.002 tonne of manure ex-housing per tonne manure ex-animal. This value is needed for the life cycle modelling of all scenarios, as data must be expressed per tonne slurry ex-animal (functional unit), but it is the slurry ex-housing which is the input into the digester.

Table S1. Reference pig slurry composition

Parameter	Slurry ex-animal ^a	Slurry ex-housing ^b	Slurry ex-storage ^c	Source and assumptions
Mass (t pig ⁻¹)	0.47	0.47 ^d	0.48	Data needed to ensure correspondence between each manure stage. Values ex-animal and ex-storage based on [4]. Value ex-housing based on mass balance ^d . A net water addition of 0.02 m ³ per tonne manure is considered during outdoor storage.
Total N (kg t ⁻¹)	6.00	5.26	5.03	N ex-animal from [4]. Losses considered (during housing and during storage): NH ₃ , N ₂ O, N ₂ , NO. Details on N losses are in Table S5. The N from straw addition ^e in-house and as a floating layer during outdoor storage is estimated as 0.009 kg per tonne manure ex-animal and 0.011 kg per tonne manure ex-storage, respectively.
P (kg t ⁻¹)	1.21	1.21	1.19	P ex-animal from [4]. No losses considered during housing and storage. The P from straw addition ^e in-house and as a floating layer during outdoor storage is estimated as 0.001 kg per tonne manure ex-animal and 0.002 kg per tonne manure ex-storage, respectively.
K (kg t ⁻¹)	2.83	2.85	2.83	K ex-animal from [4]. No losses considered during housing and storage. The K from straw addition ^e in-house and as a floating layer during outdoor storage is estimated as 0.02 kg per tonne manure ex-animal and 0.03 kg per tonne manure ex-storage, respectively.
DM (kg t ⁻¹)	74.8	68.7	66.0	DM ex-storage from [4]. Losses during storage: 5 % of the ex-housing values; losses during housing: 10 % of the ex-animal value. Assumptions for losses during storage and housing based on [5].
VS (kg t ⁻¹)	60.7	54.6	52.1	VS are assumed to constitute 79 % of the DM content. Losses considered during storage and housing (absolute values) are the same as for DM (i.e. it is assumed that all DM lost was VS).
C (kg t ⁻¹)	34.5	34.2	31.6	C ex-storage = 47.9 % of DM ex-storage for pigs, based on the ratio C: DM obtained by [6]. Losses considered (during housing and during storage): CH ₄ and CO ₂ . Details on C losses are in Table S5. The C from straw addition ^e in-house and as a floating layer during outdoor storage is estimated as 0.75 kg per tonne manure ex-animal and 0.95 kg per tonne manure ex-storage, respectively.
Cu (g t ⁻¹)	31.0	31.0	30.4	Cu ex-storage = 0.0453 % of DM ex-storage, based on the ratio Cu: DM obtained by [6]. No losses considered during housing and storage. The Cu from straw addition ^e in-house and as a floating layer during outdoor storage is estimated as 4.92 mg per tonne manure ex-animal and 6.25 mg per tonne manure ex-storage, respectively.
Zn (g t ⁻¹)	90.8	90.7	89.1	Zn ex-storage = 0.135 % of DM ex-storage, based on the ratio Zn: DM obtained by [6]. No losses considered during housing and storage. The Zn from straw addition ^e in-house and as a floating layer during outdoor storage is estimated as 75.5 mg per tonne manure ex-animal and 95.9 mg per tonne manure ex-storage, respectively.
NH ₄ -N (kg t ⁻¹)	4.20	3.94	3.07	Value ex-storage based on [4]. Value ex-housing assuming 0.75 kg NH ₄ -N per kg manure ex-housing [5], and value ex-animal assuming 0.70 kg NH ₄ -N per kg manure ex-animal [7].

^a All values of this column are expressed per tonne slurry ex-animal. ^b All values of this column are expressed per tonne slurry ex-housing. ^c All values of this column are expressed per tonne slurry ex-storage. ^d The non-rounded value ex-housing is 0.47089 t pig⁻¹, and considers a net water addition in-house of 3.57 kg water per pig, the straw addition described below and DM losses as in this Table. ^e The N, P and K addition from straw added in the stable considers, based on [5], an addition of 3 kg of straw per animal per year, 3.3 rotations per year, and the above-mentioned amount of manure ex-animal and ex-housing, yielding a total of 0.0019 t straw per tonne manure ex-housing. For the floating layer, the amount considered is based on [8], i.e. 2.5 kg per tonne manure ex-housing. The straw DM content is 85 % [9]. The N, P, K, Cu and Zn content of straw per kg of DM is 0.00528 kg, 0.0009 kg, 0.015 kg, 3 mg and 46 mg, respectively, based on [9]. The C content is taken as 0.4563 kg C per kg DM, based on an average of 13 values from the Biolex database [10].

Table S2. Life cycle inventory data for the reference manure management

Substances	Life cycle stage			Comments		
	in-house per tonne ex-animal manure	outdoor storage per tonne ex-housing manure	field per tonne ex-storage manure	in-house	outdoor storage	field
NH ₃ -N	0.71	0.099	0.60	0.17 kg NH ₃ -N per kg TAN ^a [5], with 0.7 kg TAN/kg N [7].	2.5 % of TAN ^a ex-housing [5]; the N ex-housing being estimated according to [5], i.e.: N ex-animal minus NH ₃ -N losses in-house (and not accounting for other losses).	12% of N applied [11] (this is an average for application by trail hose tanker, excluding illegal dates).
NH ₃ -N, at application			0.015			0.5% of TAN applied, for application by trail hoses, [11].
N ₂ O-N	0.012	0.030	0.050	0.002 kg N ₂ O-N per kg N ex-animal [12] (pit storage below animal)	0.005 kg N ₂ O-N per kg N ex-animal [12] (liquid/slurry storage)	1% of N applied, [13].
NO-N (representing NO _x)	1.96×10 ⁻⁴	1.84×10 ⁻⁴	0.005	0.0001 kg NO per kg TAN ex-animal [7].	0.0001 kg NO per kg TAN ex-housing [7].	0.1 × N ₂ O-N, based on [14].
NO ₃ -N	0	0	1.68	No leaching from housing, based on [1].	No leaching from outdoor storage, based on [1].	Based on Danish NLES ₄ model [15]. See section 6.
N ₂ -N	0.013	0.012		0.003 kg NO per kg TAN ex-animal [7].	0.003 kg NO per kg TAN ex-housing [7].	
CO ₂ -C	0.36	1.20	31.1	1.83 kg CO ₂ per kg CH ₄ , see section 5.	1.83 kg CO ₂ per kg CH ₄ , see section 5	Based on Danish C-TOOL model, 98.3% of the C applied end up as CO ₂ -C, over 20 y, see section 8.
CH ₄ -C	0.54	1.80	0	IPCC (2006) [12] algorithm, see Equation S1.	IPCC (2006) [12] algorithm, see Equation S1.	Assumed negligible, based on [1].
P leaching	0	0	0.060			5% of surplus, based on [16]. See section 6.
indirect N ₂ O-N (volatilization)	7.14×10 ⁻³	9.91×10 ⁻⁴	0.006	1% of N loss as NH ₃ and as NO _x , (ex-animal) [13].	1% of N loss as NH ₃ and as NO _x , (ex-housing) [13].	1% of N loss as NH ₃ and as NO _x , (ex-storage) [13].
indirect N ₂ O-N (leaching)	0	0	0.013	0.75% of N lost through leaching (ex-animal) [13].	0.75% of N lost through leaching (ex-animal) [13].	0.75% of N lost through leaching (ex-animal) [13].

^a Ammonium-N (NH₄⁺-N) and compounds readily broken down to NH₄⁺-N are referred to as total ammoniacal N (TAN).

2. System boundary for each scenario

The process flow diagrams illustrated in Figure S1-Figure S7 present the system boundary considered for each scenario. The flows indicated in the figures are related to the functional unit, i.e. 1 tonne of manure ex-animal used for anaerobic digestion.

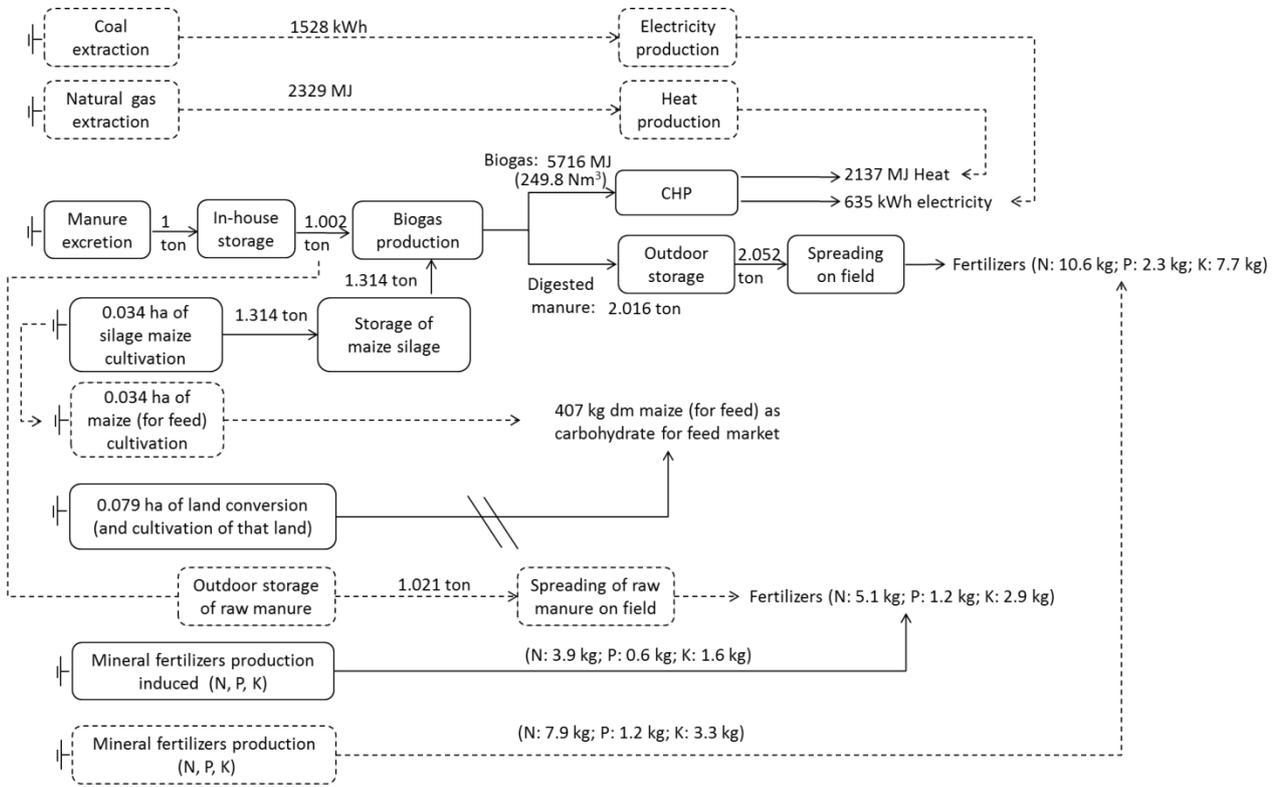


Figure S1. Process flow diagram for the maize scenario. Dotted lines indicate avoided flows. It should be noted that not all the converted land will be cultivated in feed maize, and that not all the maize (for feed) displaced is replaced, due to various market interactions.

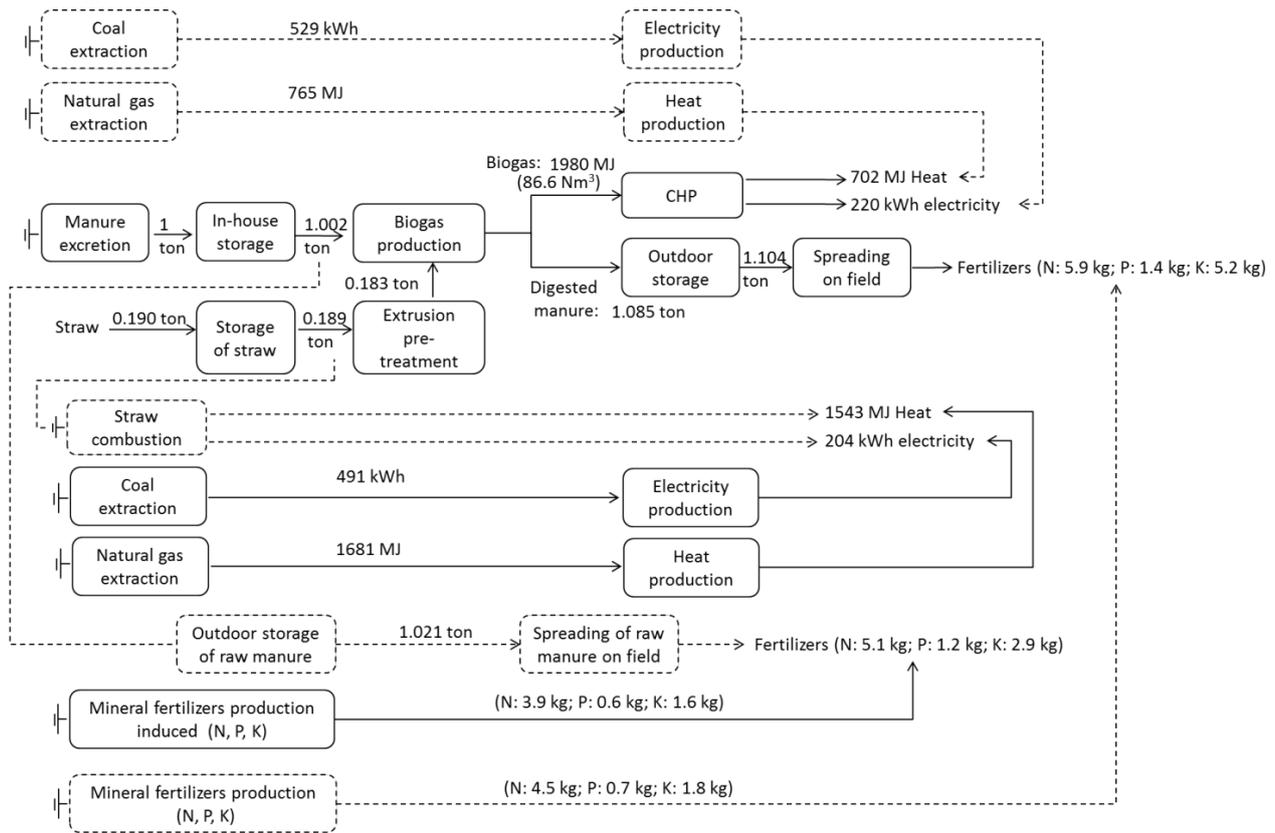


Figure S2. Process flow diagram for the straw scenario. Dotted lines indicate avoided flows.

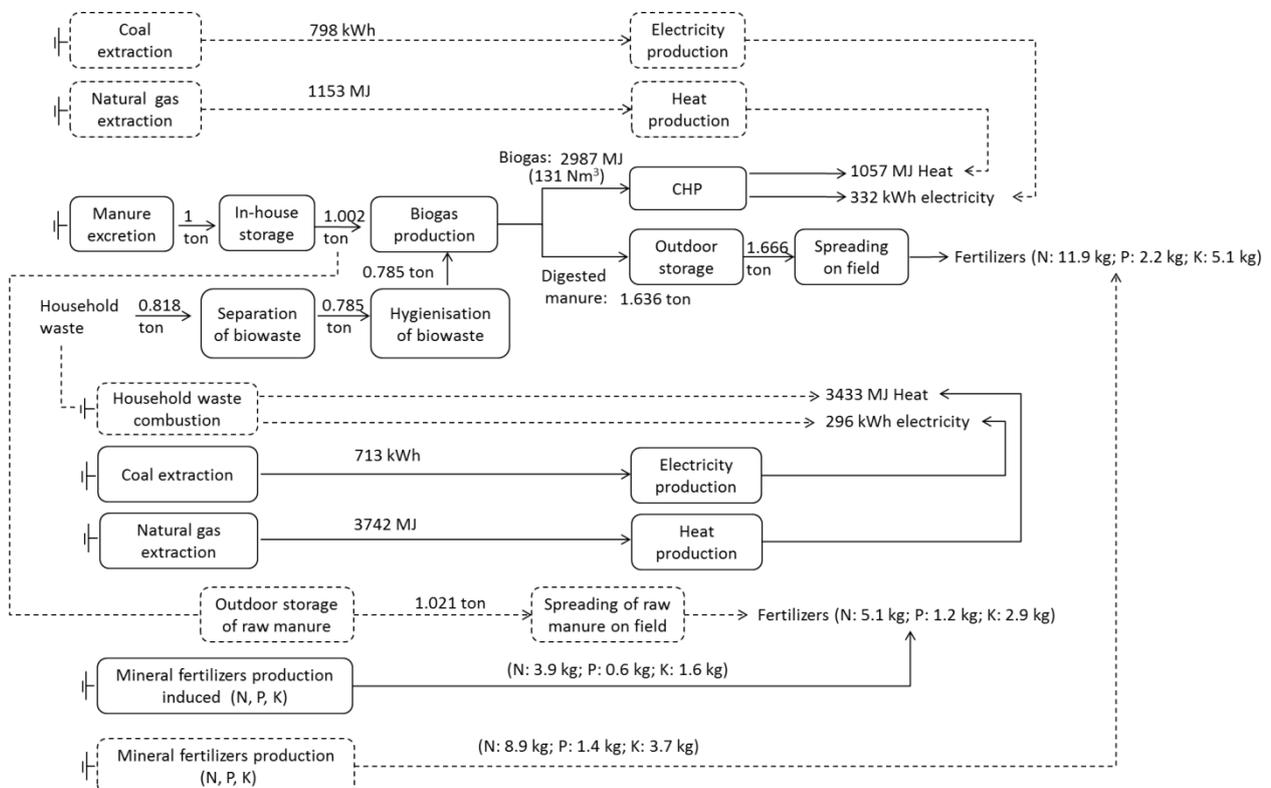


Figure S3. Process flow diagram for the household biowaste scenario. Dotted lines indicate avoided flows.

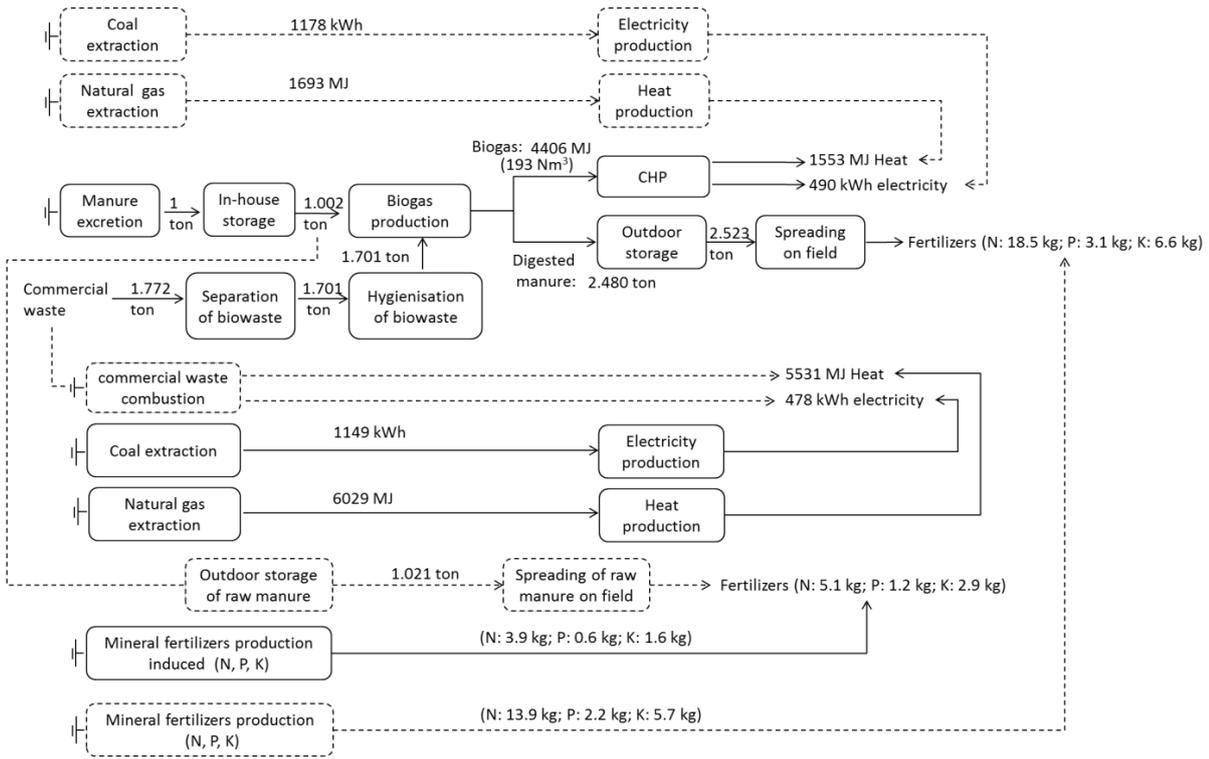


Figure S4. Process flow diagram for the commercial biowaste scenario. Dotted lines indicate avoided flows.

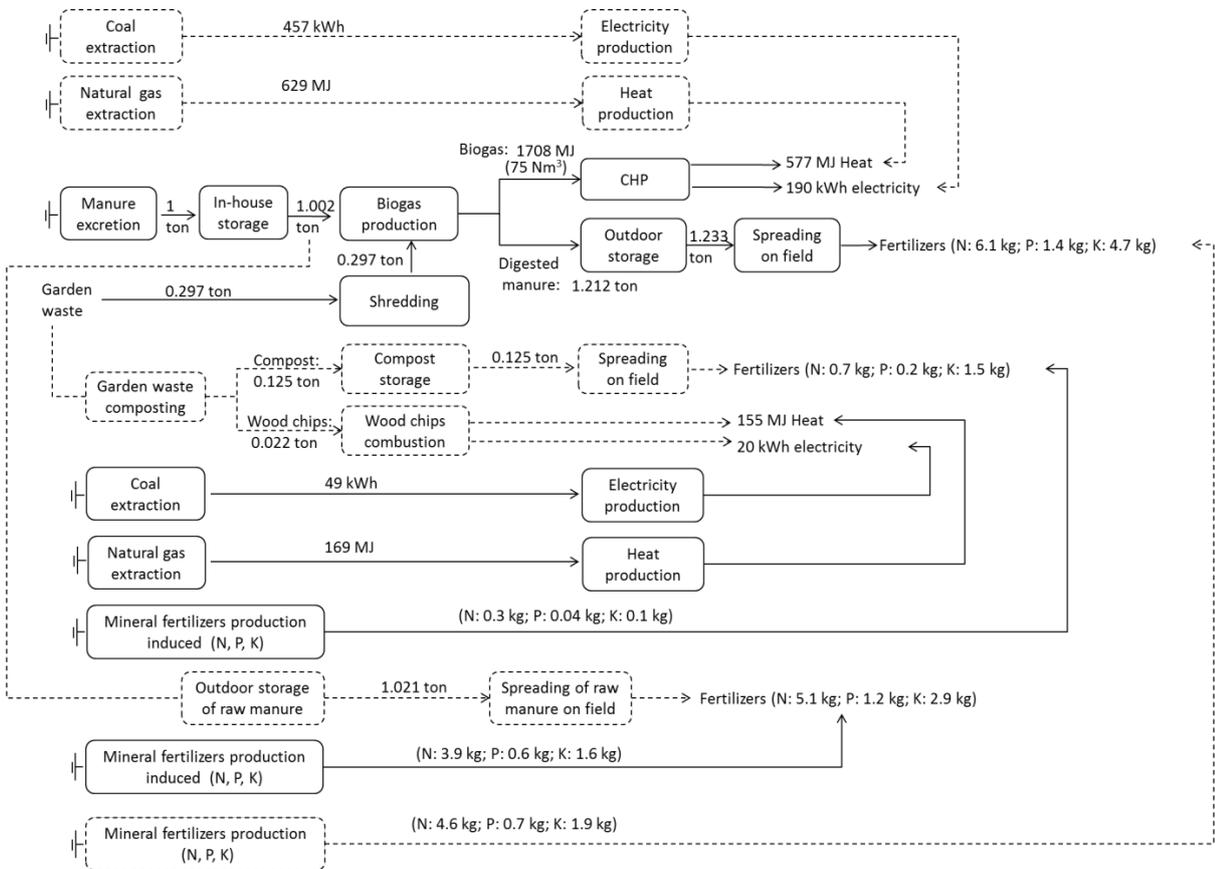


Figure S5. Process flow diagram for the garden waste scenario. Dotted lines indicate avoided flows.

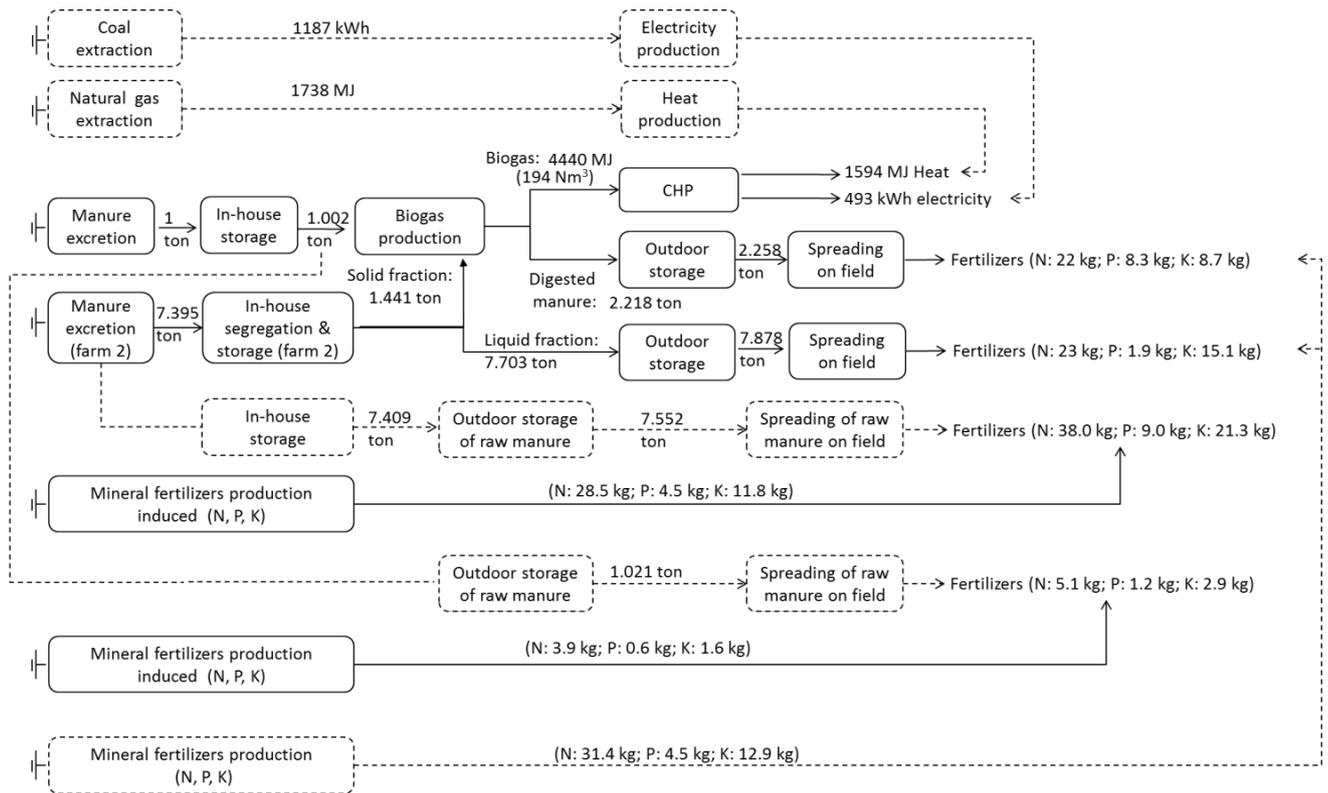


Figure S6. Process flow diagram for the source-segregation scenario. Dotted lines indicate avoided flows.

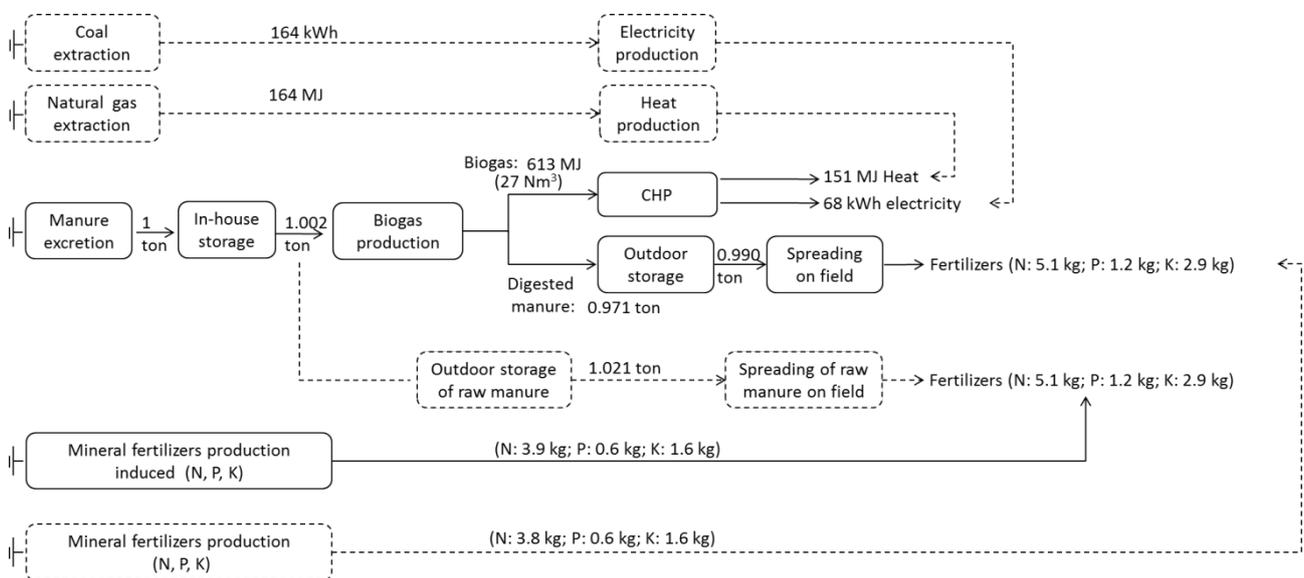


Figure S7. Process flow diagram for the mono-digestion scenario. Dotted lines indicate avoided flows.

3. Life cycle inventory

The detailed inventory is presented for each scenario. Full details are presented for the maize scenario, which serves as a reference for the other scenarios. Details on mineral fertilizer substitution are presented in section 6.

3.1 Maize scenario

3.1.1 In-house storage of manure (applies to all scenarios)

This process consists of the storage of manure inside the animal house, i.e. from the moment it is excreted (slurry ex-housing), until it is pumped towards a pre-tank. The storage duration inside animal house is assumed to 15 days, based on [17]. Emissions are as shown in Table S5. Methane emissions are based on the methodology described in the IPCC guidelines [12]:

$$\text{CH}_4 \text{ [kg]} = \text{VS [kg]} \times B_0 \times 0.67 \text{ [kg CH}_4 \text{ per m}^3 \text{ CH}_4\text{]} \times \text{MCF} \quad (\text{Equation S1})$$

Where:

VS : The amount of volatile solids as excreted by the animals (kg)

B_0 : The maximum CH_4 producing capacity for a given manure ($\text{m}^3 \text{ CH}_4 \text{ kg}^{-1} \text{ VS excreted}$) (methane potential)

MCF : Methane conversion factor (%)

The MCF factor is defined in the Revised 1996 IPCC [18] guidelines in chapter 4 (on page 4.9) as follows: “*Methane Conversion Factor (MCF)*: The MCF defines the portion of the methane producing potential (B_0) that is achieved. The MCF varies with the manner in which the manure is managed and the climate, and can theoretically range from 0 to 100 per cent. Manure managed as a liquid under hot conditions promotes methane formation and emissions. These manure management conditions have high MCFs, of 65 to 90 per cent. Manure managed as dry material in cold climates does not readily produce methane, and consequently has an MCF of about 1 per cent. Laboratory measurements were used to estimate MCFs for the major manure management techniques.” In [12], default MCF values are presented for different manure management system and in function of the average annual temperature.

For the in-house storage process, a MCF value of 3% is used, based on the default IPCC values (pit storage below animal confinement, less than 1 month, temperature $\leq 10^\circ\text{C}$). Table S3 shows the overall life cycle inventory considered for this process.

Table S3. Life cycle inventory for process “in-house storage of manure”

Comments		
Input		
Manure "ex animal"	1 000.0 kg	The input to this process is 1 000.0 kg manure “ex-animal”, which is also the study’s functional unit. The emissions are calculated relative to this.
Output		
Manure "ex housing"	1 002 kg	1 000.0 kg manure “ex animal” * (0.47089 t manure ex-housing pig ⁻¹ /0.47000 t manure ex-animal pig ⁻¹) = 1 001.9 kg manure “ex housing”. (See Table S1)
Energy consumption		
	Not included	The energy consumption for the housing units is not included within the system boundary.
Emission to air		
Carbon dioxide (CO ₂)	0.27 kg	1.83 kg CO ₂ per kg CH ₄ , see section 5
Methane (CH ₄)	0.54 kg	Based on IPCC Tier 2 approach [12] (see Equation S1) with MCF = 3 % and B ₀ of 0.40 kg CH ₄ /kg VS (based on an average of Danish data: [17,19,20]). For the calculation of B ₀ in kg CH ₄ (instead of m ³), a density of 0.717 kg CH ₄ per Nm ³ was used (so the density at 0°C, i.e. Normal conditions, was used instead of IPCC’s density at 20°C).
Ammonia (NH ₃ -N)	0.713 kg	See Table S2.
Direct emissions of Nitrous oxide (N ₂ O-N)	0.0120 kg	See Table S2.
Indirect emissions of Nitrous oxide (N ₂ O-N)	0.00713 kg	See Table S2.
Nitrogen monoxide (NO-N) (representing total NO _x)	0.000196 kg	See Table S2.
Nitrogen dioxide (NO ₂ -N)	No data	No data
Nitrogen (N ₂ -N)	0.0126 kg	See Table S2.
Discharge to water		
	None	Assumed to be zero, as leakages from housing systems are prohibited in Denmark.
Discharge to soil		
	None	Assumed to be zero, as leakages from housing systems are prohibited in Denmark.

3.1.2 Storage of manure ex-housing in pre-tank (applies to all scenarios)

This process reflects the storage of the manure “ex housing” in the pre-tank until it is pumped out in order to be delivered to the biogas plant. No significant losses (i.e. emissions) as well as no water addition are assumed for the process. Thus, the manure composition and manure quantity of the process output is assumed to be the same as the manure input, i.e. the manure ex-housing.

3.1.3 Maize cultivation and composition (maize scenario)

Maize silage is considered to be cultivated on a sandy soil (soil JB3 of the Danish soil classification) under a wet climate (precipitation: 964 mm per year). The life cycle inventory data considered for this are those described in [16], which are related to 1 ha of maize silage cultivation. These data consider a yield of 12.05 t DM/ha*y.

The maize composition of the maize silage, once harvested, is presented in Table S4 .

Table S4. Composition of maize silage, as harvested

Maize silage, as harvested	
Unit	kg/1 000.0 kg maize silage "as harvested"
DM	310.0 ^a
VS	294.5 ^c
Total N	4.31 ^a
Phosphorus(P)	0.81 ^a
Potassium (K)	3.72 ^a
Carbon (C)	139.50 ^b
Copper (Cu)	0.002 ^a
Zinc (Zn)	0.022 ^a

^a Based on [9], where all values are given as a function of the DM content (except the DM itself);

^b Based on [16], 0.45 kg C/kg DM;

^c Taken as 95 % of DM, according to [21,22].

3.1.4 Storage of maize silage at biogas plant (maize scenario)

Maize is produced during the productive season only, but can be stored and used when needed. In this study, a storage period of at least 4 months is assumed. The changes in the maize silage composition occurring during storage are based on [23]. The life cycle inventory data considered for this are presented in Table S5, and the mass balances used to calculate the composition of maize silage after storage can be found in Table S6.

Table S5. Life cycle inventory for process "storage of maize silage"

Comments		
Input		
Maize silage ("as harvested")	1 000.0 kg	The process is related to 1 000.0 kg maize silage "as harvested" (Table S4). The emissions are calculated relative to this.
Output		
Maize silage ("ex-storage")	992.0 kg	According to [23], the weight loss in storage corresponds to 0.8 % of the fresh weight: 1 000.0 kg maize silage - 0.8 % * 1 000.0 kg maize silage.
Emission to air		
Carbon dioxide (CO ₂)	5.00 kg	Taken as 0.5 % of maize silage fresh weight, based on [23].
Methane (CH ₄)		Negligible, based on [23].

Table S6. Mass balances for the maize silage storage process

	Maize silage "as delivered" ^a	Mass balance: Change during storage	Mass balance: Amount after storage	Maize silage "ex-storage" ^f
Unit	kg/1 000.0 kg maize silage "as delivered"	kg	kg	kg/1 000.0 kg maize silage "ex-storage"
Total mass	1 000.0	- 8.0 ^b	992.0	1 000.0
DM	310.0	- 6.0 ^b	304.0	306.5
VS	294.5	- 6.0 ^c	288.5	290.8
Total N	4.31	No change	4.31	4.34
Phosphorus (P)	0.81	No change	0.81	0.81
Potassium (K)	3.72	No change	3.72	3.75
Carbon (C)	139.50	- 1.36 ^d	138.14	139.3
Copper (Cu)	0.002	No change	0.002	0.002
Zinc (Zn)	0.022	No change	0.022	0.022

^a All the data are the same as given for the maize silage "as harvested" in Table S4;

^b Based on [23].

^c Assumed to be the same change as for DM;

^d This loss corresponds to the CO₂-C (Table S5): 5.00 kg CO₂ * (12/44) = 1.36 kg C lost;

^f All the data are the same as in the precedent column, but adjusted to be expressed per 1 000.0 kg of maize silage "ex-storage".

3.1.5 Biogas production from manure and maize silage (maize scenario)

As described in the main manuscript, the biogas production considered in this study is based on a two-step anaerobic digestion operated at mesophilic temperatures (around 37 °C). It is assumed that the biogas produced is constituted of 65 % CH₄ and 35% CO₂ with a density of 1.158 kg/Nm³.

The input to the anaerobic digester considered in this scenario is a biomass mixture constituted of manure "ex-housing" and maize silage "ex-storage". The amount of both fractions in the mixture entering the digester was calculated in order to get a mixture with 10% TS after the first digestion step, and with a C/N ratio of maximum 20, as also detailed in the main manuscript. The procedure for calculating this is detailed in [1]. Based on the VS biodegradability (Table S7), composition (Table S1; Table S6) and methane yields considered for maize (382 Nm³ per t VS) and manure (319 Nm³ per t), the biomass mixture was calculated to consist of 43.46% manure (ex-housing) and 56.54% maize (ex-storage).

Table S7. VS biodegradability of maize and manure

Digestion step	Manure: % of initial VS degraded	Maize silage: % of initial VS degraded
Step 1	60 ^c	79 ^c
Step 2 (including step 1)	66 ^a	87 ^b

^a Taken according to [1];

^b Taken according to [20].

^c Corresponds to 90 % of degradability at step 2.

The composition of the mixture entering the digester is established as shown in Table S8.

Table S8. Composition of the biomass mixture entering the digester for the maize scenario

	Manure "ex- housing" ^a	Maize silage "ex-storage" ^b	Mass balance: amount in manure ^c	Mass balance: amount in maize silage ^d	Biomass mixture entering digester ^e
Unit	kg / 1 000.0 kg manure	kg / 1 000.0 kg maize silage	kg / 434.6 kg manure "ex housing"	kg / 565.4 kg maize silage	kg / 1 000.0 kg biomass mixture
DM	68.7	306.5	29.9	173.3	203.1
VS	54.6	290.8	23.7	164.4	188.1
Total N	5.26	4.34	2.29	2.46	4.74
Phosphorus (P)	1.21	0.81	0.53	0.46	0.99
Potassium (K)	2.85	3.75	1.24	2.12	3.36
Carbon (C)	34.25	139.25	14.89	78.73	93.61
Copper (Cu)	0.031	0.002	0.013	0.001	0.014
Zinc (Zn)	0.091	0.022	0.039	0.013	0.052

^a All the data are the same as given for the manure "ex-housing" in Table S1;

^b All the data are the same as given for the maize silage "ex-storage" in Table S6;

^c All the data are the same as in the column "Manure "ex-housing", but adjusted to be expressed per 434.6 kg of manure (the share of manure in the 1 000.0 kg biomass mixture input);

^d All the data are the same as in the column "Maize silage", but adjusted to be expressed per 565.4 kg maize silage (the share of maize silage in the 1 000.0 kg biomass mixture input);

^e Calculated as the sum (i.e. column "Mass balance: amount in manure" + column "Mass balance: amount in maize silage").

The amount of biogas produced from 1 tonne of biomass mixture is thus of 108.4 Nm³ biogas, calculated as:

- Biogas from manure fraction: $434.6 \text{ kg manure "ex-housing"} * 54.6 \text{ kg VS}/1\,000.0 \text{ kg manure "ex-housing"} * 319 \text{ Nm}^3 \text{ CH}_4/\text{t VS} / 0.65 \text{ Nm}^3 \text{ CH}_4/\text{Nm}^3 \text{ biogas} * \text{t}/1\,000 \text{ kg} = 11.6 \text{ Nm}^3 \text{ biogas}$;
- Biogas from maize silage fraction: $565.4 \text{ kg maize silage} * 290.8 \text{ kg VS}/1\,000.0 \text{ kg maize silage "ex-storage"} * 382 \text{ Nm}^3 \text{ CH}_4/\text{t VS} / 0.65 \text{ Nm}^3 \text{ CH}_4/\text{Nm}^3 \text{ biogas} * \text{t}/1\,000 \text{ kg} = 96.7 \text{ Nm}^3 \text{ biogas}$;

The life cycle inventory data for the biogas production process are presented in Table S9.

Table S9. Life cycle inventory data for process “biogas production”, for the maize scenario

Comments		
Input		
Biomass mixture (manure + maize silage)	1 000.0 kg	1 000.0 kg of the biomass mixture constituted as detailed in text.
Output		
Biogas (65 % CH ₄ and 35 % CO ₂)	125.5 kg (108.4 Nm ³)	$108.4 \text{ Nm}^3 \text{ biogas} * 1.158 \text{ kg biogas/Nm}^3 \text{ biogas (biogas density)} = 125.5 \text{ kg biogas.}$
Digestate	874.5 kg	No water loss. Therefore, the only loss is the mass of the biogas: $1\,000.0 \text{ kg biomass mixture} - 125.5 \text{ kg biogas.}$
Energy consumption		
Electricity	14.00 kWh	Estimated own consumption of electricity: 5 % of net production, engine efficiency of 40 % (see main manuscript). Electricity from the grid.
Heat	110.57 MJ	Heating the biomass from 8 °C to 37 °C, assuming that the specific heat for DM corresponds to 3.00 kJ/kg °C and to 4.20 kJ/kg °C for water.
Emission to air		
Carbon dioxide (CO ₂)	1.126 kg	Calculated as a function of the CH ₄ emissions, assuming a ratio of 2.23 kg CO ₂ per kg CH ₄ (see section 8).
Methane (CH ₄)	0.505 kg	1% of the methane content of the biogas is assumed to be emitted to the environment (see main manuscript). $108.4 \text{ Nm}^3 \text{ biogas} * 65 \% \text{ CH}_4 * 0.717 \text{ kg CH}_4/\text{Nm}^3 \text{ CH}_4 * 1 \% = 0.505 \text{ kg CH}_4$, with methane (CH ₄) density of 0.717 kg/Nm ³
Odour		No data.
Discharge to water and soil		
	None	No emissions are considered.

No water loss is assumed to happen in the digester. Thus, the total input mass, DM and VS loss corresponds to the mass of biogas produced only. Based on [1], it is considered that there are no losses of N, P, and K occurring during the digestion. The C losses correspond to the sum of CO₂-C and CH₄-C transferred to the biogas, and to the emissions (fugitive losses) of CO₂-C and CH₄-C occurring during the digestion process.

Table S10 shows the mass balance considered in order to calculate the composition of the digestate as it leaves the digester.

Table S10. Mass balance for the biomass mixture before and after the anaerobic digestion, maize scenario

	Biomass mixture entering digester ^a	Mass balance: Change during biogas production	Mass balance: Amount after biogas production	Digestate "ex-digester" ^f
Unit	kg/1 000.0 kg biomass mixture	kg	kg	kg/1 000.0 kg digestate
Total mass	1 000.0	- 125.5 ^b	874.5	1000
DM	203.1	- 125.5 ^c	77.6	88.8
VS	188.1	- 125.5 ^d	62.6	71.6
Total N	4.74	No change	4.74	5.42
Phosphorus (P)	0.99	No change	0.99	1.13
Potassium (K)	3.36	No change	3.36	3.84
Carbon (C)	93.61	- 58.97 ^e	34.64	39.61
Copper (Cu)	0.014	No change	0.014	0.016
Zinc (Zn)	0.052	No change	0.052	0.059

^a All the data are the same as given for the biomass mixture entering digester in Table S8.

^b This loss corresponds to the biogas produced, expressed in mass terms (see Table S9);

^c No water loss and therefore change in dry matter is equal to change in total mass;

^d The same change as for DM (all the DM loss was VS);

^e This corresponds to the losses in the biogas itself and the losses that occurred during the digestion process:

- losses in the biogas are calculated as the sum of CH₄-C and CO₂-C: $(108.4 \text{ Nm}^3 \text{ biogas} * 65 \% \text{ CH}_4 * 0.717 \text{ kg CH}_4/\text{Nm}^3 \text{ CH}_4) * (12.011 \text{ g/mol} / 16.04 \text{ g/mol}) + (108.4 \text{ Nm}^3 \text{ biogas} * 35 \% \text{ CO}_2 * 1.977 \text{ kg CO}_2/\text{Nm}^3) * (12.011 \text{ g/mol} / 44.01 \text{ g/mol}) = 58.29 \text{ kg C}$, where 0.717 kg CH₄/Nm³ CH₄ is the methane density, and 1.977 kg CO₂/Nm³ CO₂ is the carbon dioxide density.
- losses from the digestion process are the aggregated losses as CO₂-C + CH₄-C: $1.126 \text{ kg CO}_2 * (12.011 \text{ g/mol} / 44.01 \text{ g/mol}) + 0.505 \text{ kg CH}_4 * (12.011 \text{ g/mol} / 16.04 \text{ g/mol}) = 0.69 \text{ kg C}$, with CO₂ and CH₄ emissions taken from Table S9. The total loss: 58.29 kg C + 0.69 kg C = 58.97 kg C.

^f All the data are the same as in the precedent column, but adjusted to be expressed per 1000 kg of digestate.

3.1.6 Co-generation of heat and power from biogas (applies to all scenarios)

The produced biogas is burned in a biogas engine, as detailed in the main manuscript. Table S11 shows the emissions related to this process.

Table S11. Life cycle inventory for process “co-generation of heat and power from biogas”

Comments		
Input		
Biogas (65 % CH ₄ and 35 % CO ₂)	0.044 Nm ³ (1 MJ)	Amount of biogas corresponding to an energy content of 1 MJ: $1 \text{ MJ}/22.88 \text{ MJ/Nm}^3 = 0.044 \text{ Nm}^3$ (22.88 MJ/Nm ³ is the biogas heat value, see main manuscript)
Output		
Heat	0.46 MJ	The heat efficiency of the biogas engine is 46 %, see manuscript.
Electricity	0.40 MJ	The electricity efficiency of the biogas engine is 40 %, see manuscript.
Emission to air		
Carbon dioxide (CO ₂)	8.36×10 ⁻²	Reference [24], table 34.
Carbon monoxide (CO)	3.10×10 ⁻⁴	Reference [25], table 19.
Methane (CH ₄)	4.34×10 ⁻⁴	Reference [25], table 19.
Non-methane volatile organic compounds (NMVOC)	1.00×10 ⁻⁵	Reference [25], table 19.
Ammonia (NH ₃)		No data
Nitrous oxide (N ₂ O)	1.60×10 ⁻⁶	Reference [25], table 19.
Nitrogen oxides (NO _x)	2.02×10 ⁻⁴	Reference [25], table 19.
Nitrogen monoxide (NO)		No data
Nitrogen(N ₂)		No data
Particulates		
PM ₁₀	4.51×10 ⁻⁷	Reference [24], table 65.
PM _{2.5}	2.06×10 ⁻⁷	Reference [24], table 65.
Hydrogen sulphide (H ₂ S)		No data
Sulphur dioxide (SO ₂)	1.92×10 ⁻⁵	Reference [25], table 33.
Odour		No data
Discharge to water		
	None	No emissions to water
Discharge to soil		
	None	No emissions to soil

3.1.7 Avoided heat production (maize scenario)

It is assumed in this study that the biogas plant is connected to the district heating grid, and that natural gas is the marginal heat source displaced. The Ecoinvent process “Heat, natural gas, at boiler atmospheric low-NO_x non-modulating <100kW/RER U” was used to model this (described in [26]: table 13.9, p. 161). In reality, displacing heat from a CHP plant also has incidence on the electricity, and this should also be modelled. For simplifying the model, this was however not included in this study.

Table S12 presents the net heat avoided per functional unit (FU).

Table S12. Net heat production (maize scenario)

Parameter	Value	Comment
Biogas produced, per tonne mixture	108.4 Nm ³	Table S9
Biogas produced per FU	249.8 Nm ³	1) The amount of biomass mixture per FU is: 1.002 t manure ex-housing/FU (Table S3)/ 434.6 kg manure ex-housing/t biomass mixture (Table S8) = 2.305 t mixture/FU. 2) The amount of biogas per FU thus corresponds to: 108.4 Nm ³ /t mixture * 2.305 t mixture/FU = 249.8 Nm ³ biogas per FU.
Total heat produced, per FU	2629.4 MJ	249.8 Nm ³ biogas per FU * 46% heat efficiency * 22.9 MJ/Nm ³ biogas = 2624 MJ heat/FU
Heat needed for process, per FU	254.9 MJ	110.57 MJ/t mixture (Table S9) * 2.305 t mixture/FU = 254.9 MJ heat/FU
Net avoided heat, per FU	2137.0 MJ	2629.4 MJ – 254.9 MJ * 90% heat used (see manuscript) = 2137.0 MJ

3.1.8 Avoided electricity production (maize scenario)

The electricity produced from the biogas is displacing coal-based electricity (assumed as marginal electricity for Denmark). This was modelled through the Ecoinvent process “Electricity, hard coal, at power plant/NORDEL U” (described in [27]: table 11.14, p. 226).

Table S13 presents the net avoided electricity.

Table S13. Net electricity avoided, per FU

Parameter	Value	Comment
Biogas produced, per tonne mixture	108.4 Nm ³	Table S9
Biogas produced per FU	249.8 Nm ³	Table S12
Total electricity produced, per FU	2686.4 MJ	249.8 Nm ³ biogas per FU * 40% electricity efficiency * 22.9 MJ/Nm ³ biogas = 2286 MJ heat/FU
Net avoided electricity, per FU	635.1 kWh	2686.4 MJ/3.6 MJ per kWh

3.1.9 Storage of digestate (maize scenario)

Table S14 presents the life cycle inventory for the process “digestate storage”.

Table S14. Life cycle inventory for process “digestate storage, maize scenario”

Comments		
Input		
Digestate ("ex-digester")	1 000.0 kg	The process is related to the 1 000.0 kg digestate "ex biogas plant". The emissions are calculated relative to this.
Water	20.0 kg	The water from precipitation. Fixed to 20 kg rain/1 000.0 kg biomass (taken identical to water addition during the outdoor storage of manure, see Table S1).
Straw layer	2.5 kg	The life cycle data of straw production are not included in this study, as being regarded as a waste product from cereal production.
Output		
Digestate ("ex-storage")	1018.2 kg	Digestate leaving the storage. The composition change due to the addition of rainwater, and to the losses (emissions).
Energy consumption		
Electricity	2.90 kWh	Electricity for pumping and stirring the digestate (based on [8]).
Emission to air		
Carbon dioxide (CO ₂)	2.941 kg	2.23 kg CO ₂ /kg CH ₄ as established in see section 5.
Methane (CH ₄)	1.319 kg	Calculated with IPCC guidelines (see Equation S1) using MCF = 10 % and B ₀ = 0.458 m ³ CH ₄ /kg VS (calculated for this mixture, based on the proportion and methane yield of each fraction of the mixture). To this, an “emission reduction potential” factor of 50 % is applied, accounting for the lower emissions of digestates [28]: 71.6 kg VS/t digestate (Table S10) * 0.458 m ³ CH ₄ /kg VS * 0.67 kg CH ₄ /m ³ CH ₄ * 10 % * (100 - 50) % = 1.319 kg CH ₄ .
Ammonia (NH ₃ -N)	0.122 kg	NH ₃ -N = 2.5 % of the TAN (as in Table S2), assuming TAN = 90 % of total N ^a : 2.5 % * 85 % * 5.42 kg N/1 000.0 kg digestate = 0.122 kg NH ₃ -N.
Direct emissions of Nitrous oxide (N ₂ O-N)	0.0271 kg	Calculated using an emission factor of 0.005 kg N ₂ O-N per kg N (as in Table S2): 0.005 N ₂ O-N per total N in digestate "ex-biogas plant" * 5.42 kg N/1 000.0 kg digestate * (100 - 40) % = 0.0271 kg N ₂ O-N.
Indirect emissions of Nitrous oxide (N ₂ O-N)	0.00122 kg	Indirect emissions due to the volatilization of NH ₃ and NO _x : 0.01 kg N ₂ O-N per kg (NH ₃ -N + NO _x -N) volatilized (as in Table S2).
Nitrogen monoxide (NO-N) (representing total NO _x)	0.000228 kg	NO = 0.0001 of the TAN (as in Table S2), with TAN = 90 % of the total N ^a : 5.42 kg N/1 000.0 kg digestate * 0.0001 kg NO-N/kg TAN * 90 % * (14/30) = 0.000228 kg NO-N.
Nitrogen dioxide (NO ₂ -N)	No data	No data
Nitrogen (N ₂ -N)	0.01464 kg	N ₂ -N = 0.003 of the TAN (as in Table S2), with TAN = 90 % of the total N ^a : 5.42 kg N/1 000.0 kg digestate * 0.003 kg N ₂ -N/kg TAN * 90 % = 0.01464 kg N ₂ -N.
Discharge to water		
	None	Assumed (leakage from storage tank prohibited in Denmark).

^a Estimated, assuming 100 % of the total N is TAN for maize silage degassed (based on [29]) and 77 % of the N is TAN for the degassed fattening manure (taken according to [11]): 100 % * 565.4 kg maize silage/1 000.0 kg biomass mixture + 77 % * 434.6 kg manure “ex-housing”/1 000.0 kg biomass mixture = 90 %.

Based on Table S14, the mass balances can be established in order to determine the composition of the digestate ex-storage, as shown in Table S15.

Table S15. Mass balances for the digestate storage process, maize scenario

	Digestate "ex-digester" ^a	Mass balance: Change during storage of digestate	Mass balance: Amount after storage of digestate	Digestate "ex-storage" ^f
Unit	kg/1000 kg digestate"ex-digester "	kg	kg	kg/1000 kg digestate"ex-storage"
Total mass	1000	18.2 ^b	1 018.2	1 000.0
DM	88.8	- 1.82 ^c	86.9	85.4
VS	71.6	- 1.82 ^c	69.8	68.6
Total N	5.42	- 0.16 ^d	5.26	5.16
Phosphorus (P)	1.13	No change	1.13	1.11
Potassium (K)	3.84	No change	3.84	3.77
Carbon (C)	39.61	- 1.79 ^e	37.82	37.15
Copper (Cu)	0.016	No change	0.016	0.016
Zinc (Zn)	0.059	No change	0.059	0.058

^a From Table S10.

^b Equals to the mass of water added during the storage minus DM loss;

^c The change is calculated as the sum of N and C losses (see below); the authors acknowledge that the value is underestimated, but this rough estimation was made as this value is not used for further estimations;

^d Changes in total N equal to the sum of N losses due to N-emissions occurring during the digestate storage:

$0.122 \text{ kg } \text{NH}_3\text{-N} + 0.0271 \text{ kg } \text{N}_2\text{O-N (direct)} + 0.000228 \text{ kg } \text{NO-N} + 0.01464 \text{ kg } \text{N}_2\text{-N} = 0.16 \text{ kg N};$

^e Changes in total C equal to the sum of C losses due to CO₂ and CH₄ emissions occurring during the digestate storage: $2.941 \text{ kg } \text{CO}_2 * (12.011 \text{ g/mol}/44.01 \text{ g/mol}) + 1.319 \text{ kg } \text{CH}_4 * (12.011 \text{ g/mol}) / 16.04 \text{ g/mol} = 1.66 \text{ kg C};$

^f All the data are the same as in the precedent column, but adjusted to be expressed per 1 000.0 kg of digestate "ex-storage".

3.1.10 Spreading of digestate (maize scenario)

The process "digestate spreading on land" reflects the field application of the digestate mixture. Application by trail hoses tanker is assumed. The life cycle inventory for this process is shown in Table S16.

Table S16. Life cycle data for process “digestate spreading on land”, maize scenario

Comments		
Input		
Digestate “ex storage”	1 000.0 kg	The process is related to 1 000.0 kg digestate "ex-storage". The emissions occurring during the process are calculated relative to this.
Output		
Digestate on field, fertilizer value		See section 6.
Emission to air		
Carbon dioxide (CO ₂)	96.2	Based on C-TOOL, 71 % of the C applied ends up as CO ₂ , considering a 20 y annualization (section 8).
Methane (CH ₄)	Negligible	Assumed to be negligible, as the formation of CH ₄ requires anoxic environment, and the field is aerobic.
Ammonia (NH ₃ -N) during application	0.0238 kg	0.5 % of TAN "ex-storage" (as in Table S2), with the TAN “ex-storage” being evaluated as 92 % of total N ^a . calculated as: $5.16 \text{ kg N} * 92 \% * 0.5 \% = 0.0238 \text{ kg NH}_3\text{-N}$.
Ammonia (NH ₃ -N) in period after application	0.620 kg	0.12 kg NH ₃ -N per kg total N in the degassed biomass (as in Table S2): $5.16 \text{ kg N} * 0.12 \text{ kg NH}_3\text{-N/kg total N} = 0.62 \text{ kg NH}_3\text{-N}$.
Direct emissions of Nitrous oxide (N ₂ O-N)	0.0310 kg	Based on IPCC guidelines, correspond to 0.01 kg N ₂ O-N per kg N applied (as in Table S2): $5.16 \text{ kg N} * 0.01 \text{ kg N}_2\text{O-N per kg N} = 0.031 \text{ kg N}_2\text{O-N}$.
Indirect emissions of Nitrous oxide (N ₂ O-N)	0.00647 kg 0.0129 kg	Indirect emissions due to volatilization of ammonia (NH ₃ -N) and nitrogen oxides (NO _x -N). Based on IPCC guidelines, correspond to 0.01 kg N ₂ O-N per kg (NH ₃ -N + NO _x -N) volatilized (as in Table S2). Indirect emissions due to nitrate leaching. Based on IPCC guidelines, correspond to 0.0075 kg N ₂ O-N per kg N leaching.
Nitrogen oxides (NO _x -N)	0.00310 kg	NO _x -N = 0.1 * N ₂ O-N (direct), (as in Table S2).
Discharge to water		
Nitrate leaching	1.726 kg N	Nitrate leaching to the water bodies. Based on N-LES ₄ model [15], see section 7.
Phosphorus leaching	0.0555 kg P	Phosphorus leaching reaching the water recipients, see section 7.
Discharge to soil		
Copper (Cu)	0.0161 kg	100% of the Cu in the digestate applied, based on [8].
Zinc (Zn)	0.0584 kg	100% of the Zn in the digestate applied, based on [8].

^a Taken as for digestate “ex-digester” (90 %), considering an increase of 2 % due to storage (based on [11]).

3.1.11 Direct land use changes

Direct land use changes represent the change resulting from using the land for cultivating the maize needed for this biogas scenario instead of using it for what it would have been used for instead. In a country like Denmark, where 65% 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) [3], 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 [30].

Based on findings from [31], spring barley is often designated as the marginal crop for Western European countries [e.g. 32–34]. However, this scenario looks towards the long-term and aims to reflect a high bioenergy future in which the demand for biomass and arable land has increased to a large extent. In such a future it is seen as likely that the benefits of the greater (and potentially increased) yield of maize as compared to barley has changed the cropping towards maize for both animal feed and energy. Based on this, the additional hectares of maize needed for anaerobic co-digestion is modeled to displace the hectares of maize used for feed (as the other feed crops would already be phased out and imported). As the production of maize silage (for energy) instead of maize (for feed), which represents the direct land use change (DLUC) involved in this study, was assumed to result in negligible changes in emissions, the DLUC was excluded from the model (but considered in sensitivity analyses, see section 9.4).

3.1.12 Indirect land use change (ILUC) (maize scenario)

The first step in the calculation of the indirect land use change (ILUC) impact is knowing the amount of land displaced in Denmark because maize (for biogas) is now cultivated. As detailed above, it was assumed that maize (for feed) is the crop “kicked-out” for cultivating the maize-for-biogas. There is 1303 kg maize silage “ex-storage” needed per FU (565.4 kg maize needed per t mixture × 2.305 t mixture per FU, Table S12), which on the basis of Table S6, corresponds to 1313.8 kg maize silage “as harvested” per FU. Based on the maize DM content (310 kg DM/t maize “as harvested”, Table S6), and on the maize yield (taken at 12.05 t DM ha⁻¹, based on [16]), 0.0338 ha of Danish land are required to cultivate the amount of maize needed per FU (this holds no matter the crop displaced). In the baseline case, this means that feed maize corresponding to 0.0338 ha, i.e. 0.407 ton DM of feed maize, is no longer provided to the market.

This drop of supply will trigger an increase in the price for carbohydrates feedstock, which then provides incentives to increase the production elsewhere [35–37]. 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) [35–37]. This study includes the latter only.

In order to get a link between the amount of displaced feed maize and the amount of land converted to agriculture, the recent PhD work of Kløverpris [36] was used. Using a modified version of the Global Trade Analysis Project (GTAP) model, Kløverpris [36], modeled the ILUC resulting from a marginal increase in wheat consumption in 4 different countries, including Denmark. Kløverpris results show how much land is converted, for different biomes of the world, due to 1 tonne of wheat demand increase from Denmark. These results have been used as a proxy to estimate the ILUC impact involved in this study (Table S17).

Kløverpris results [36] indicate an expansion of 0.1658 ha of land per extra tonne of wheat demanded, which corresponds to 0.1950 ha of land per extra ton of wheat DM demanded (considering wheat has a DM content of 85%, based on [9]). Considering the “ton wheat DM demanded” as a proxy for “ton DM demanded of a carbohydrate crop”, this 0.1950 ha expanded/ton DM demanded figure was multiplied by the yield of the displaced maize, i.e. 12.05 ton DM/ha. This results in a figure of 2.35 ha of land expanded per ha of feed maize displaced.

In Table S17, the results in terms of how many ha will be converted, and for which biomes of the world, are presented, based on Kløverpris's results, as above-described. The CO₂ impact of land conversion was however not estimated by Kløverpris. In order to do so, the soil and vegetation C data from the Woods Hole Research Centre, as published in [35], have been used, and the CO₂ emitted due to land conversion was calculated based on the methodology published in [38]. Based on this methodology, it was considered that 25% of the C in the soil is released as CO₂ for all types of land use conversion, except when forests were converted to grassland, where 0% of the C is released. Further, it was considered that 100% of the C in vegetation is released as CO₂ for all forest types as well as for tropical grassland conversions, while 0% is released for the remaining biome types (e.g. shrub land, non-tropical grassland, chaparral). Details are presented in Table S17. It should be noted that this approach is exactly the same as used in a parallel publication [30]. In [30], however, an adjustment is considered to take into account the yield of the crop displaced. In this study, the yield is not considered for the GHG effect, but only for estimating the land expanded.

The uncertainties level in Table S17 are based on the qualitative "certainty evaluation" performed by Kløverpris on his own results (m² expanded per tonne wheat) [36]. An uncertainty of 20% has been considered in this study for Kløverpris's level "very good", 40% for the level "good", 60% for the level "moderate", and 80% for the level "poor".

Table S17 presents an ILUC figure of 357 t CO₂ eq. per ha displaced. If annualized over 20 years (as in the Renewable Energy Directive, for example), this corresponds to 17.85 t CO₂ eq. per ha per y. This is the figure that was used in this study (in the main manuscript, this figure is presented with two significant digits, i.e. 18 t CO₂ eq. per ha per y).

Table S17. ILUC impact^a

Biomes converted ^b	Type of conversion ^c	Region ^{c,d}	m ² t ⁻¹ wheat ^{c,e}	C in vegetation (t ha ⁻¹) ^f	C in soil (t ha ⁻¹) ^f	CO ₂ -C lost (t C t ⁻¹ wheat) ^g	CO ₂ lost (t CO ₂ t ⁻¹ wheat)	CO ₂ lost (t CO ₂ ha displaced ⁻¹) ^h
Savanna (taken as shrub land)	100% cropland	xss	140 ± 86	4.6	30	0.11 ± 0.06	0.39 ± 0.24	2.59 ± 1.55
African tropical evergreen forest (taken as tropical rain forest)	100% cropland	xss	140 ± 86	130	190	2.5 ± 1.5	9.1 ± 5.5	60.10 ± 36.06
Open shrubland (taken as shrub land)	100% grassland	xss	81 ± 49	4.6	30	0.06 ± 0.04	0.22 ± 0.13	1.48 ± 0.89
Temperate evergreen forest	100% cropland	xeu15	57 ± 34	160	130	1.1 ± 0.7	4.0 ± 2.4	26.66 ± 16.00
Temperate deciduous forest	100% cropland	xeu15	57 ± 34	120	130	0.87 ± 0.52	3.2 ± 1.9	21.15 ± 12.69
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	28.27 ± 16.96
Tropical evergreen forest	100% cropland	bra	180 ± 70	200	98	4.0 ± 1.6	15 ± 6	95.75 ± 38.30
Savanna (taken as grassland)	100% grassland	bra	41 ± 16	10	42	0.04 ± 0.02	0.16 ± 0.06	1.05 ± 0.42
Grassland/steppe (taken as temperate grassland)	100% cropland	xsu	91 ± 55	10	190	0.43 ± 0.26	1.6 ± 0.9	10.41 ± 6.25
Temperate evergreen forest	100% grassland	xsu	45 ± 27	160	130	0.88 ± 0.43	3.2 ± 1.6	17.54 ± 10.53
Temperate deciduous forest	100% grassland	xsu	45 ± 27	140	130	0.76 ± 0.37	2.8 ± 1.3	14.80 ± 8.88
Savanna (taken as tropical grassland)	100% cropland	aus	110 ± 64	18	42	0.31 ± 0.18	1.1 ± 0.7	7.39 ± 4.43
Open shrubland + grassland/steppe (taken as tropical grassland)	100% grassland	aus	37 ± 22	18	42	0.11 ± 0.06	0.39 ± 0.23	2.55 ± 1.53
Boreal deciduous forest (taken as temperate deciduous forest)	100% cropland	can	97 ± 58	140	130	1.6 ± 1.0	6.0 ± 3.6	39.50 ± 23.70
Boreal evergreen forest (taken as temperate evergreen forest)	100% grassland	can	10 ± 6	160	130	0.16 ± 0.10	0.59 ± 0.35	3.87 ± 2.32
Grassland/steppe (taken as grassland)	100% cropland	xla	35 ± 21	10	42	0.04 ± 0.02	0.14 ± 0.08	0.90 ± 0.54
Tropical evergreen forest	100% cropland	xla	35 ± 21	200	98	0.79 ± 0.48	2.9 ± 1.7	19.17 ± 11.50
Savanna + dense shrub land (taken as grassland)	100% grassland	xla	16 ± 10	10	42	0.02 ± 0.01	0.063 ± 0.038	0.42 ± 0.25
Open shrub land (taken as chaparral)	100% grassland	usa	68 ± 41	40	80	0.14 ± 0.08	0.50 ± 0.30	3.29 ± 1.97
TOTAL	-	-	1500 ± 880	-	-	15 ± 8	54 ± 30	357 ± 195

a Eventual inconsistencies due to rounding

b Indicated biomes are as in [36]. When the biomes mentioned in [36] did not figure in the biomes from the Woods Hole Research Centre data [35], an equivalent was considered, which is indicated between parentheses, when it applies.

c Based on the results from [36].

d 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 [36], this aggregation covers 92% of the total net expansion.

e The maximal and minimal range are based on the qualitative description of the uncertainty related to the biomes conversion results made by [36]. The levels identified as “very good”, “good” and “moderate” were considered as an uncertainty of $\pm 20\%$, 40% and 60%, respectively.

f From the Woods Hole Research Centre, as published in [35].

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

h The conversion per ha is made considering a wheat yield of $5.61 \text{ t DM ha}^{-1}$ and a DM content of 85% of the crop fresh matter.

3.1.13 Reacting crop production (maize scenario)

The above hectares of land expanded are afterwards cultivated, based on the rationale described in section 5.1.11. As these are converted as a result of displaced Danish feed maize, it can be expected that a carbohydrate crop (e.g. wheat, maize, rice, barley, sorghum, millet, rye and oats) will be cultivated in these newly available hectares of arable land. For each of the main regions where expansion takes place (Table S17), the reacting carbohydrate crop (among wheat, maize, rice, barley, sorghum, millet, rye and oats) has been identified. This has been done based on the FAO statistics on the quantity produced of these crops (in each region), for the years 2001-2010. For each region, there was typically one of the carbohydrate crops that have been much more produced than the other over this time period. This crop was then designated as the “reacting crop”. When two crops had close production volumes, the one with the greatest increase was considered as the “reacting crop”.

Table S18 shows the “reacting carbohydrate crop” for each of the region where expansion takes place.

Table S18. Reacting crop mix

Region	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	Total
Net expansion ^a , m ²	367	361	217	182	144	107	87	68	125	1 658
Reacting crop ^b	Maize	Wheat	Maize	Wheat	Wheat	Barley	Maize	Wheat	-	
Country ^b	Botswana	France	Brazil	Kazakhstan	Australia	Canada	Argentina	United States of America	-	
Share ^c (%)	22 %	22 %	13 %	11 %	9 %	6 %	5 %	4 %	8 %	100 %
Share, adjusted ^d (%)	24 %	24 %	14 %	12 %	9 %	7 %	6 %	4 %	-	100 %

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

^a Net expansion resulting from 1 tonne extra wheat demand from Denmark (results from [36]) . These results differ slightly from Table S18 as that Table only presents 2 significant digits.

^b Defined using FAOSTAT statistic bank for crop production [39];

^c Calculated as the share of the total area to be converted, f.ex.: *area to be converted in Brazil [ha] / total area to be converted*;

^d The data is the same as in the row above, but adjusted by excluding the share of “Rest of the world”.

The life cycle data considered for each reacting crop are presented in Table S19.

Table S19. Life cycle data for reacting crop production

	Yield, t (fresh weight) / ha	Contribution to 1 ha net expansion		Process used in modeling
		ha reacting crop / 1 ha net expansion ^c	t (fresh weight) / 1 ha net expansion ^d	
Reacting crop production		1 ha	3.1573 t	The process is related to 1 ha land cultivation.
Maize in Botswana	0.22 ^a	0.2391	0.0519	Ecoinvent process for corn cultivation in USA "Corn, at farm/US U".
Wheat in France	6.95 ^b	0.2356	1.6366	Ecoinvent process for wheat cultivation in France: "Wheat grains conventional, Barrois, at farm/FR U".
Maize in Brazil	3.59 ^a	0.1419	0.5099	Ecoinvent process for corn cultivation in USA: "Corn, at farm/US U".
Wheat in Kazakhstan	1.04 ^a	0.1185	0.1234	Ecoinvent process for wheat cultivation in Spain: "Wheat grains conventional, Castilla-y-Leon, at farm/kg/ES".
Wheat in Australia	1.55 ^a	0.0940	0.1453	Ecoinvent process for wheat cultivation in Spain: "Wheat grains conventional, Castilla-y-Leon, at farm/kg/ES".
Barley in Canada	2.80 ^b	0.0697	0.2066	The process of barley cultivation in Canada modeled based on [40].
Maize in Argentina	6.52 ^a	0.0568	0.3700	Ecoinvent process for corn cultivation in USA: "Corn, at farm/US U".
Wheat in United States of America	2.82 ^b	0.0443	0.1249	Ecoinvent process for wheat cultivation in USA: "Wheat grains, at farm/US U".

^a Corresponds to the average yield according FAOSTAT statistics bank [39], assessed for the period of 2001 – 2010;

^b Taken according to the Ecoinvent process used in modeling;

^c Contribution to 1 ha net expansion in terms of area (ha); calculated as, f.ex. for maize in Botswana: 24 % * 1 ha, where 24 % is the share of Botswana's maize considered to contribute to the reacting crop mix (see Table S18);

^d Contribution to 1 ha net expansion in terms of crop mass (t); calculated from the two preceding columns: yield (t/ha) * specific area cultivated (ha/1 ha net expansion);

3.1.14 Reference manure handling avoided: raw manure outdoor storage (applies for all scenarios)

This process consists of the outdoor storage of raw "ex-housing" manure in a concrete tank, which is covered by a floating layer of straw (2.5 kg straw per tonne of manure ex-housing) [1]. The process described here (Table S20) is identical as in the one in [1], but is based on the manure composition for this study (Table S1). Further, the ratio CO₂/CH₄ considered here is different (see section 5).

Table S20. Life cycle data for process “outdoor storage of raw manure”

Input		Comments
Manure ("ex housing")	1 000.0 kg	The process is related to 1000.0 kg manure "ex-housing". The emissions are calculated relative to this.
Water	20 kg	The water from precipitation: 0.02 m ³ per tonne manure (Table S1).
Straw floating layer	2.5 kg	Based on [1]. The life cycle data of straw production are not included in this study, as being regarded as a waste product from cereal production.
Output		
Manure ("ex storage")	1018.0 kg	With composition as shown in Table S1.
Energy consumption		
Electricity	2.90 kg	Electricity for pumping and stirring of raw manure, based on [8].
Emission to air		
Carbon dioxide (CO ₂)	4.39 kg	See Table S2.
Methane (CH ₄)	2.40 kg	Calculated with IPCC guidelines (Equation S1) using MCF = 10 % and B ₀ = 0.40 kg CH ₄ /kg VS ^a and VS of manure "ex housing" (Table S1): 54.6 kg VS/t manure * 0.40 m ³ CH ₄ /kg VS * 0.67 kg CH ₄ /m ³ CH ₄ * 10 % = 2.40 kg CH ₄ .
Ammonia (NH ₃ -N)	0.099 kg	See Table S2.
Direct emissions of Nitrous oxide (N ₂ O-N)	0.0299 kg	See Table S2.
Indirect emissions of Nitrous oxide (N ₂ O-N)	0.00099 kg	See Table S2.
Nitrogen monoxide (NO-N) (representing total NO _x)	0.00018 kg	See Table S2.
Nitrogen (N ₂ -N)	0.0118 kg	See Table S2.
Discharge to water		
	None	Assumed to be zero.

^a From an average of Danish values: 0.38, 0.35, 0.36 and 0.5 m³ CH₄/kg VS (12, 13, 31). For the calculation of B₀ in kg CH₄ (instead of m³), a density of 0.717 kg CH₄ per Nm³ was used (so the density at 0°C, i.e. Normal conditions, was used instead of IPCC's density at 20°C).

3.1.15 Reference manure handling avoided: raw manure spreading (applies for all scenarios)

This process consists of the application of the manure ex-storage on land, through a trail hoses slurry tanker. The inventory data used for this process are exactly as presented in Table S2.

3.2 Straw scenario

3.2.1 Harvest of straw and straw composition (straw scenario)

The straw reference used in this study is represented by the winter wheat straw (as being the most abundant in Denmark) with a yield of 3.09 t DM per ha [16], a methane potential of 432 Nm³ CH₄ / kg VS and a degradability of 45 % [19]. The harvest process involves swath, baling and loading (of the bales), and these were modeled as described in [16]. The straw composition considered is shown in Table S21.

Table S21. Straw composition

Unit	Straw "as harvested" kg/1 000.0 kg straw "as harvested"
Total mass	1 000.0
DM	850.0 ^a
VS	810.6 ^c
Total N	4.49 ^a
Phosphorus (P)	0.77 ^a
Potassium (K)	12.75 ^a
Carbon (C)	382.50 ^b
Copper (Cu)	0.003 ^a
Zinc (Zn)	0.039 ^a

^a Based on [9];

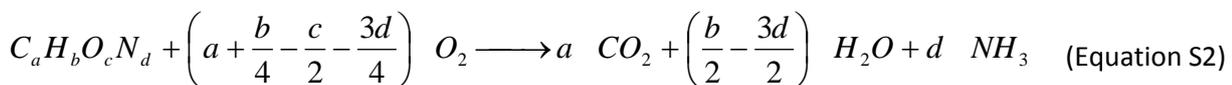
^b Based on [41], 0.45 kg C/kg DM;

^c Taken as 95 % of DM, according to [19,20,42].

3.2.2 Storage of straw

Once produced, straw bales are assumed to be stored, until used for biogas. A storage period longer than 4 months is considered.

The changes in the composition of straw during the storage are modeled based on [43], where indoor storage of straw dry bales during 8 months was investigated and a loss of 1.1 % of the initial DM was observed. Based on Equation S2 (from [44]), but corrected for the sign for the term (3d/4) for O₂ describing the aerobic degradation of organic matter (for a complete conversion), all the DM loss occurring during the storage is assumed to be in form of carbon dioxide (CO₂) and ammonia (NH₃) emissions.



Based on this equation, the ratio between CO₂ and NH₃ to be produced can be calculated (Table S22).

Table S22. Calculation of ratio between CO₂ and NH₃ for straw aerobic degradation, during storage

Organic component	Parameter	Value	CO ₂	NH ₃
VS lipid (1 mol) (C ₅₇ H ₁₀₄ O ₆)	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS lipid (Equation S2)		57	-
	Weight (%) for VS lipid in straw (see section 5) (g lipid/g straw)	1.63%		
	Lipid molecular weight	884 g/mol		
	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS lipid, per kg straw		1.649	-
VS protein (1 mol) (C ₅ H ₇ O ₂ N)	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS protein (Equation S2)		5	1
	Weight (%) for VS protein in straw (see section 5) (g protein/g straw)	2.66%		
	Protein molecular weight	113 g/mol		
	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS protein, per kg straw		1.846	0.369
VS VFA (1 mol) (C ₂ H ₄ O ₂)	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS VFA (Equation S2)		2	-
	Weight (%) for VS VFA in straw (see section 5) (g VFA/g straw)	0.00%		
	VFA molecular weight	60 g/mol		
	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS VFA, per kg straw		0.00	0.00
VS carbohydrates easily degradable (1 mol) (C ₆ H ₁₀ O ₅)	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS carbohydrates easily degradable (Equation S2)		6	-
	Weight (%) for VS carbohydrates easily degradable in straw (see section 5) (g carbohydrates/g straw)	54.42%		
	Carbohydrates molecular weight	162 g/mol		
	Moles of CO ₂ and NH ₃ from the degradation of 1 mole VS carbohydrates easily degradable, per kg straw		20.156	-
SUM (moles of CO ₂ and NH ₃ per kg straw)			23.651	0.369
Ratio CO₂/NH₃		64 moles CO₂ per mole NH₃ (165.9 g CO₂/g NH₃)		

Based on the above, the life cycle inventory for straw storage could be established (Table S23).

Table S23. Life cycle inventory for storage of straw

Comments		
Input		
Straw ("as harvested")	1 000.0 kg	The process is related to the 1000.0 kg straw as harvested.
Output		
Straw ("ex-storage")	990.7 kg	No water loss. Therefore, the only loss is the DM loss occurring due to the biological degradation of the easy degradable VS in the straw. This loss is taken as 1.1 % of the initial DM, based on [43].
Emission to air		
Carbon dioxide (CO ₂)	15.43 kg	Based on the assumption that all C lost during the storage is CO ₂ -C: 4.21 kg C lost (from Table S24) * (44/12) = 15.43 kg CO ₂ .
Ammonia (NH ₃ -N)	0.077 kg	Calculated as a function of the CO ₂ emissions (Table S22): 165.9 g CO ₂ per g NH ₃ .

The mass balance related to this storage process is presented in Table S24.

Table S24. Mass balance for the straw before and after storage

	Straw "as harvested"	Mass balance: Change during storage	Mass balance: Amount after storage	Straw "ex-storage"
Unit	kg/1 000.0 kg straw "as harvested"	kg	kg	kg/1 000.0 kg straw "ex-storage"
Total mass	1 000.0	-9.4 ^b	990.7	1 000.0
DM	850.0	-9.4 ^a	840.7	848.6
VS	810.6	-9.4 ^b	801.3	808.8
Total N	4.49	-0.08 ^c	4.41	4.45
Phosphorus (P)	0.77	No change	0.77	0.77
Potassium (K)	12.75	No change	12.75	12.87
Carbon (C)	382.50	-4.21 ^d	378.29	381.86
Copper (Cu)	0.003	No change	0.003	0.003
Zinc (Zn)	0.039	No change	0.039	0.039

^a Based on [43], assumed to be the 1.1 % initial DM;

^b Assumed the same as for DM;

^c Ammonia losses

^d Calculated proportional to the total dry matter loss: 9.4 kg DM lost * 382.50 kg C/1 000.0 kg straw "as harvested" / 850.0 kg DM/1 000.0 kg straw "as harvested" = 4.21 kg C lost;

3.2.3 Extrusion pre-treatment (straw scenario)

This process was modeled based on the results from [45], and all details are available in the main manuscript. The energy consumption for the pre-treatment (14.5 kWh per t of straw) consists of 7.5 kWh of electricity for cutting the straw (based on [46]), and 7.0 kWh of electricity consumed to operate the extruder [45]. The mass balance for this process is as shown in Table S25.

Table S25. Mass balance for the straw before and after the extrusion pre-treatment

	Straw "ex-storage"	Mass balance: Change during extrusion	Mass balance: Amount after extrusion	Straw "extruded"
Unit	kg/1 000.0 kg straw "ex storage"	kg	kg	kg/1 000.0 kg straw "extruded"
Total mass	1000.0	-30.0 ^b	970.0	1000.0
DM	848.6	-25.5 ^a	823.1	848.6
VS	808.8	-24.3 ^b	784.6	808.8
Total N	4.45	-0.13 ^b	4.32	4.45
Phosphorus (P)	0.77	-0.02 ^b	0.75	0.77
Potassium (K)	12.87	-0.39 ^b	12.48	12.87
Carbon (C)	381.86	-11.46 ^b	370.41	381.86
Copper (Cu)	0.003	-0.0001 ^b	0.002	0.003
Zinc (Zn)	0.039	-0.001 ^b	0.038	0.039

^a According to [45], corresponds to 3 % DM initial;

^b Calculated proportional to the DM loss;

3.2.4 Biogas production

Here, the same considerations as described in section 3.1.5 for the maize silage scenario apply. Based on the VS biodegradability (Table S26), composition and methane yields considered for extruded straw (263 Nm³ per t VS) and manure (319 Nm³ per tonne), the biomass mixture was calculated to consist of 84.56% manure (ex-housing) and 15.44% straw (ex-extrusion).

Table S26. VS biodegradability, straw scenario

Digestion step	Manure: % of initial VS degraded	Extruded straw: % of initial VS degraded
Step 1	60 ^c	55 ^c
Step 2 (including step 1)	66 ^a	61 ^b

^a Taken according to [1].

^b As detailed in the main manuscript, straw degradability is considered as 45%, and to this, an increase of 35% is considered due to the extrusion pre-treatment (45% + (45%×35%).

^c Corresponds to 90 % of degradability at step 2.

The composition of the mixture entering the digester and mass balances are shown in Table S27.

Table S27. Composition of the biomass mixture for the straw scenario (manure “ex-housing” + straw “ex-trusion”).

	Manure ("ex-housing")	Straw ("extruded")	Mass balance: amount in manure	Mass balance: amount in straw	Biomass mixture entering digester
Unit	kg / 1 000.0 kg manure “ex housing”	kg / 1 000.0 kg straw “ex treatment”	kg / 845.6 kg manure “ex housing”	kg / 154.4 kg straw “extruded”	kg / 1 000.0 kg biomass mixture
DM	68.7	848.6	58.1	131.0	189.1
VS	54.6	808.8	46.1	124.9	171.0
Total N	5.26	4.45	4.45	0.69	5.14
Phosphorus (P)	1.21	0.77	1.02	0.12	1.14
Potassium (K)	2.85	12.87	2.41	1.99	4.40
Carbon (C)	34.25	381.86	28.96	58.96	87.92
Copper (Cu)	0.031	0.003	0.026	0.0004	0.027
Zinc (Zn)	0.091	0.039	0.077	0.006	0.083

The emissions due to the anaerobic digestion process, as well as the heat and electricity consumed were modelled using the same algorithms as described in Table S9, but the CO₂/CH₄ ratio (for calculating the biogenic CO₂ releases) was here considered as 1.94 (see section 5). As a result of the biogas production, the composition of the digestate, as it leaves the digester, will be different as the composition of mixture that entered the digester. This is shown in Table S28.

Table S28. Mass balance for the biomass mixture (straw scenario) before and after the anaerobic digestion process.

	Biomass mixture entering digester	Mass balance: Change during biogas production	Mass balance: Amount after biogas production	Digestate “ex biogas plant”
Unit	kg/1 000.0 kg biomass mixture	kg	kg	kg/1 000.0 kg digestate
Total mass	1 000.0	- 84.6 ^a	915.4	1 000.0
DM	189.1	- 84.6 ^b	104.5	114.2
VS	171.0	- 84.6 ^c	86.4	94.4
Total N	5.14	No change	5.14	5.61
Phosphorus (P)	1.14	No change	1.14	1.25
Potassium (K)	4.40	No change	4.40	4.80
Carbon (C)	87.92	- 39.72 ^d	48.20	52.65
Copper (Cu)	0.027	No change	0.027	0.029
Zinc (Zn)	0.083	No change	0.083	0.090

^a This loss corresponds to the biogas produced (73.1 Nm³ biogas per t mixture), expressed in mass terms (density of 1.158 kg Nm⁻³);

^b No water loss and therefore change in dry matter is equal to change in total mass;

^c The same change as for DM (all DM lost was VS);

^d This corresponds to the losses in the biogas itself and the losses that occurred during the digestion process; estimated as described in Table S10.

3.2.5 Co-generation of heat and power (straw scenario)

This process was modeled as described in section 3.1.6.

3.2.6 Avoided heat production (straw scenario)

This process was modeled as described in section 3.1.7.

3.2.7 Avoided electricity production (straw scenario)

This process was modeled as described in section 3.1.8.

3.2.8 Storage of the digestate (straw scenario)

For this process, the life cycle inventory is based on the same algorithms as shown in Table S14, except that a value of 1.94 is considered for the ratio CO₂: CH₄. The B₀ calculated for the mixture (based on the methane potential of each substrate, and their proportion in the mixture) was 0.475 m³ CH₄/kg VS. Further, it was here considered that 77% of the digestate's N is TAN (considering that 77% of the N is TAN for both fractions). Table S29 presents the mass balance related to this process.

Table S29. Mass balance of the digestate (straw scenario) before and after storage

	Digestate "ex biogas plant"	Mass balance: Change during storage of digestate	Mass balance: Amount after storage of digestate	Digestate "ex-storage"
Unit	kg/1000 kg digestate "ex biogas plant"	kg	kg	kg/1000 kg digestate "ex storage"
Total mass	1 000.0	17.6 ^a	1017.6	1000
DM	114.2	- 2.45 ^b	111.7	109.8
VS	94.4	- 2.45 ^c	92.0	90.4
Total N	5.61	- 0.14 ^d	5.47	5.37
Phosphorus (P)	1.25	No change	1.25	1.23
Potassium (K)	4.80	No change	4.80	4.72
Carbon (C)	52.65	- 2.30 ^d	50.35	49.48
Copper (Cu)	0.029	No change	0.029	0.029
Zinc (Zn)	0.090	No change	0.090	0.089

^a Same water addition as for the maize scenario, minus DM loss (given below);

^b The change is calculated as a sum of N and C losses; the authors acknowledge that the value is underestimated, but this rough assumption was allowed as the value is irrelevant in the further life cycle stages;

^c Estimated as equal to DM (all DM losses were VS);

^d Estimated as in the maize scenario;

3.2.9 Digestate spreading on land (straw scenario)

This process was modeled as described in Table S16.

3.2.10 Avoided straw combustion (straw scenario)

As described in the main manuscript, if straw would not have been used for anaerobic digestion, it is considered that it would have been used for combustion in a small-to-medium scale biomass CHP plant with efficiencies of 27% and 63% for electricity and heat, respectively. For this process, the composition of straw as it leaves the storage (straw ex-storage; Table S24) is considered. The LHV of straw is taken as

16.999 MJ per kg straw DM [47], which corresponds to 14.425 MJ/kg straw wet weight. The life cycle inventory for this process is presented in Table S30.

Table S30. Life cycle inventory for the avoided straw combustion process

Comments		
Input		
Straw "ex-storage"	0.069 kg	Amount of straw corresponding to an energy content of 1 MJ input: $1 \text{ MJ}/14.425 \text{ MJ/kg straw} = 0.069 \text{ kg straw}$.
Output		
Heat	0.27 MJ	The efficiency of the heat production at biomass CPH plant.
Electricity	0.63 MJ	The efficiency of the electricity production at biomass CPH plant.
Emission to air		
Carbon dioxide (CO ₂)	9.71 E-02 kg	Estimated as the difference between the total carbon in straw (ex-storage) and the CH ₄ -C loss (given below): $(0.069 \text{ kg straw "ex storage"} * 381.86 \text{ kg C}/1 \text{ 000.0 kg straw "ex storage"} - 4.40 * 10^{-3} * 12/16) * 44/12 = 0.0971 \text{ kg CO}_2$.
Carbon dioxide (CO ₂), fossil	4.40 E-03 kg	Due to the fossil fuel consumption needed in order to run the process. Taken as for biowaste incineration, but adjusted to be expressed per straw quantity, i.e. $0.069 \text{ kg straw} * 63.4 \text{ kg CO}_2 / 1 \text{ 000.0 kg biomass incinerated} = 4.40 * 10^{-3} \text{ kg CO}_2$.
Sulphur dioxide (SO ₂)	4.9E-05 kg	Reference [25]
Nitrogen oxides (NO _x)	1.25E-04 kg	Reference [25]
Non-methane volatile organic compounds (NMVOC)	7.80E-07 kg	Reference [25]
Methane (CH ₄)	4.70E-07 kg	Reference [25]
Nitrous oxide (N ₂ O)	1.1E-06 kg	Reference [25]
TSP	2.30E-06 kg	Reference [25]
Cd	3.20E-10 kg	Reference [25]
Hg	3.10E-10 kg	Reference [25]
Zn	4.1E-10 kg	Reference [25]
PCDD/-F ^a	1.90E-14 kg	Reference [25]
PAH (BaP)	1.25E-10 kg	Reference [25]
ΣPAH	5.95E-09 kg	Reference [25]
Naphthalene	1,21E-08 kg	Reference [25]
HCB	1,10E-13 kg	Reference [25]
HCl	5.60E-05 kg	Reference [25]
Discharge to water and soil		
	None	No emissions to water and soil are considered.

^a Modeled as "Dibenzofuran".

The induced heat and electricity produced due to the energy no longer provided by straw combustion is as shown in Figure S2.

3.2.11 Reference manure handling avoided: storage and spreading

These processes are as described in sections 3.1.14 and 3.1.15.

3.3 Household biowaste scenario

3.3.1 Biowaste separation and hygienisation

These processes are as described in the main manuscript.

3.3.2 Biogas production (household biowaste scenario)

Here, the same considerations as described in section 3.1.5 for the maize silage scenario apply. Based on the VS biodegradability (Table S31), composition and methane yields considered for household biowaste (330 Nm³ per t VS) and manure (319 Nm³ per t), the biomass mixture was calculated to consist of 56.06% manure (ex-housing) and 43.94% household biowaste (ex-hygenisation).

Table S31. VS biodegradability, household biowaste scenario

Digestion step	Manure: % of initial VS degraded	Household biowaste: % of initial VS degraded
Step 1	60 ^c	58 ^c
Step 2 (including step 1)	66 ^a	64 ^b

^a Taken according to [16].

^b As detailed in the main manuscript.

^c Corresponds to 90 % of degradability at step 2.

The composition of the mixture entering the digester and mass balances are shown in Table S32.

Table S32. Composition of the biomass mixture for the household biowaste scenario (manure “ex-housing” + household biowaste “ex-hygenisation”).

	Manure ("ex-housing")	Biowaste for biogas ^a	Mass balance: amount in manure	Mass balance: amount in biowaste	Biomass mixture entering digester
Unit	kg / 1 000.0 kg manure “ex- housing”	kg / 1 000.0 kg biowaste	kg / 560.6 kg manure “ex-housing”	kg / 439.4 kg biowaste	kg / 1 000.0 kg biomass mixture
DM	68.7	315.0	38.5	138.4	176.9
VS	54.6	259.8	30.6	114.2	144.8
Total N	5.26	8.79	2.95	3.86	6.81
Phosphorus (P)	1.21	1.29	0.68	0.57	1.25
Potassium (K)	2.85	2.80	1.60	1.23	2.83
Carbon (C)	34.25	141.75	19.20	62.29	81.49
Copper (Cu)	0.031	0.022	0.017	0.010	0.027
Zinc (Zn)	0.091	0.009	0.051	0.004	0.055

^a See Table S58.

The emissions due to the anaerobic digestion process, as well as the heat and electricity consumed were modelled using the same algorithms as described in Table S9, but the CO₂/CH₄ ratio (for calculating the biogenic CO₂ releases) was here considered as 1.80 (see section 5). As a result of the biogas production, the composition of the digestate, as it leaves the digester, will be different as the composition of mixture that entered the digester. This is shown in Table S33.

Table S33. Mass balance for the biomass mixture (household biowaste scenario) before and after the anaerobic digestion process.

	Biomass mixture entering digester	Mass balance: Change during biogas production	Mass balance: Amount after biogas production	Digestate "ex biogas plant"
Unit	kg/1 000.0 kg biomass mixture	kg	kg	kg/1 000.0 kg digestate
Total mass	1 000.0	- 84.6 ^a	915.4	1000
DM	176.9	- 84.6 ^b	92.4	100.9
VS	144.8	- 84.6 ^c	60.2	65.8
Total N	6.81	No change	6.81	7.44
Phosphorus (P)	1.25	No change	1.25	1.36
Potassium (K)	2.83	No change	2.83	3.09
Carbon (C)	81.49	- 39.70 ^d	41.79	45.65
Copper (Cu)	0.027	No change	0.027	0.030
Zinc (Zn)	0.055	No change	0.055	0.060

^a This loss corresponds to the biogas produced (73.0 Nm³ biogas per t mixture), expressed in mass terms (density of 1.158 kg Nm⁻³);

^b No water loss and therefore change in dry matter is equal to change in total mass;

^c The same change as for DM (all DM lost was VS);

^d This corresponds to the losses in the biogas itself and the losses that occurred during the digestion process; estimated as described in Table S10.

3.3.3 Co-generation of heat and power (household biowaste scenario)

This process was modeled as described in section 3.1.6.

3.3.4 Avoided heat production (household biowaste scenario)

This process was modeled as described in section 3.1.7.

3.3.5 Avoided electricity production (household biowaste scenario)

This process was modeled as described in section 3.1.8.

3.3.6 Storage of the digestate (household biowaste scenario)

For this process, the life cycle inventory is based on the same algorithms as shown in Table S14, except that a value of 1.80 is considered for the ratio CO₂: CH₄. The B₀ calculated for the mixture (based on the methane potential of each substrate, and their proportion in the mixture) was 0.499 m³ CH₄/kg VS. Further, it was here considered that 77% of the digestate's N is TAN (considering that 77% of the N is TAN for both fractions).

Table S34 presents the mass balance related to this process.

Table S34. Mass balance of the digestate (household biowaste scenario) before and after storage

	Digestate "ex biogas plant"	Mass balance: Change during storage of digestate	Mass balance: Amount after storage of digestate	Digestate "ex storage"
Unit	kg/1000 kg digestate "ex biogas plant"	Kg	kg	kg/1000 kg digestate "ex storage"
Total mass	1000	18 ^a	1018	1000
DM	100.9	-1.83 ^b	99.1	97.3
VS	65.8	-1.83 ^c	63.9	62.8
Total N	7.44	-0.19 ^d	7.25	7.12
Phosphorus (P)	1.36	No change	1.36	1.34
Potassium (K)	3.09	No change	3.09	3.04
Carbon (C)	45.65	-1.64 ^d	44.01	43.22
Copper (Cu)	0.030	No change	0.030	0.029
Zinc (Zn)	0.060	No change	0.060	0.059

^a Same water addition as for the maize scenario, minus DM loss (given below);

^b The change is calculated as a sum of N and C losses; the authors acknowledge that the value is underestimated, but this rough assumption was allowed as the value is irrelevant in the further life cycle stages;

^c Estimated as equal to DM (all DM losses were VS);

^d Estimated as in the maize scenario;

3.3.7 Digestate spreading on land (household biowaste scenario)

This process was modeled as described in Table S16.

3.3.8 Avoided biowaste combustion (household biowaste scenario)

As described in the main manuscript, if household biowaste would not have been used for anaerobic digestion, it is considered that it would have been used for combustion in a municipal solid waste incineration CHP plant, with electricity and heat efficiencies corresponding to 20.7% and 74% respectively, and a LHV of 20.00 MJ kg⁻¹ DM (main manuscript). Based on the DM content of the biowaste, this corresponds to a LHV of 6.3 MJ kg⁻¹ biowaste.

The life cycle inventory for this process is presented in Table S35.

Table S35. Life cycle inventory for the avoided combustion and CHP production from household biowaste

Comments		
Input		
Biowaste	1 000.0 kg	Related to 1 000.0 kg biowaste incineration.
Output		
Heat	0.21 MJ	The efficiency of the electricity production at the municipal solid waste incineration CHP plant.
Electricity	0.74 MJ	The efficiency of the heat production at the municipal solid waste incineration CHP plant.
Emission to air		
Sulphur dioxide (SO ₂)	7.50E-02 kg	Obtained from the EASEWASTE model [48]
Nitrogen oxides (NO _x)	1.40E+00 kg	Obtained from the EASEWASTE model [48]
Non-methane volatile organic compounds (NMVOC)	9.00E-03 kg	Obtained from the EASEWASTE model [48]
Methane (CH ₄)	3.62E-01 kg	Obtained from the EASEWASTE model [48]
Carbon monoxide (CO)	8.12E-02 kg	Obtained from the EASEWASTE model [48]
Nitrous oxide (N ₂ O)	6.58E-04 kg	Obtained from the EASEWASTE model [48]
Ammonia (NH ₃)	2.93E-06 kg	Obtained from the EASEWASTE model [48]
As	5.77E-07 kg	Obtained from the EASEWASTE model [48]
Cd	9.94E-08 kg	Obtained from the EASEWASTE model [48]
Co	4.48E-08 kg	Obtained from the EASEWASTE model [48]
Cr	2.12E-06 kg	Obtained from the EASEWASTE model [48]
Cu	1.55E-06 kg	Obtained from the EASEWASTE model [48]
Hg	6.24E-07 kg	Obtained from the EASEWASTE model [48]
Mn	2.94E-07 kg	Obtained from the EASEWASTE model [48]
Ni	3.61E-06 kg	Obtained from the EASEWASTE model [48]
Pb	8.65E-07 kg	Obtained from the EASEWASTE model [48]
Sb	5.40E-10 kg	Obtained from the EASEWASTE model [48]
Se	3.95E-06 kg	Obtained from the EASEWASTE model [48]
Ti	9.73E-11 kg	Obtained from the EASEWASTE model [48]
V	1.00E-05 kg	Obtained from the EASEWASTE model [48]
Zn	2.44E-06 kg	Obtained from the EASEWASTE model [48]
PAH (BaP)	2.82E-08 kg	Obtained from the EASEWASTE model [48]
HCl	3.00E-02 kg	Obtained from the EASEWASTE model [48]
HF	6.22E-05 kg	Obtained from the EASEWASTE model [48]
Benzene	7.02E-08 kg	Obtained from the EASEWASTE model [48]
Carbon dioxide (CO ₂), biogenic	5.20E+02 kg	Obtained from the EASEWASTE model [48]
Carbon dioxide (CO ₂), fossil	6.34E+01 kg	Obtained from the EASEWASTE model [48]
Dioxin, 2,3,7,8 Tetrachlorodibenzo-p-	3.40E-10 kg	Obtained from the EASEWASTE model [48]
Hydrocarbons, unspecified	4.66E-03 kg	Obtained from the EASEWASTE model [48]
Hydrogen sulfide (H ₂ S)	2.81E-07 kg	Obtained from the EASEWASTE model [48]
Particulates	2.13E-03 kg	Obtained from the EASEWASTE model [48]
Discharge to water		
Ammonia (NH ₃)	1.58E-10 kg	Obtained from the EASEWASTE model [48]
Ammonium (NH ₄ ⁺)	3.29E-05 kg	Obtained from the EASEWASTE model [48]
Chloride (Cl)	4.47E-01 kg	Obtained from the EASEWASTE model [48]
Sulfate (SO ₄ ⁻)	1.54E-02 kg	Obtained from the EASEWASTE model [48]
Discharge to soil		
	None	No emissions to soil are considered.

The induced heat and electricity produced due to the energy no longer provided by household biowaste combustion is as shown in Figure S3.

3.3.9 Reference manure handling avoided: storage and spreading

These processes are as described in sections 3.1.14 and 3.1.15.

3.4 Commercial biowaste scenario

3.4.1 Biowaste separation and hygienisation

These processes are as described in the main manuscript.

3.4.2 Biogas production (commercial biowaste scenario)

Here, the same considerations as described in section 3.1.5 for the maize silage scenario apply. Based on the VS biodegradability (Table S36), composition and methane yields considered for commercial biowaste (277 Nm³ per t VS) and manure (319 Nm³ per t), the biomass mixture was calculated to consist of 37.07% manure (ex-housing) and 62.93% commercial biowaste (ex-hygienisation). The composition of the mixture entering the digester and mass balances are shown in Table S37.

Table S36. VS biodegradability, commercial biowaste scenario

Digestion step	Manure: % of initial VS degraded	Commercial biowaste: % of initial VS degraded
Step 1	60 ^c	52 ^c
Step 2 (including step 1)	66 ^a	57 ^b

^a Taken according to [1].

^b Methane yield/Methane potential.

^c Corresponds to 90 % of degradability at step 2.

Table S37. Composition of the biomass mixture for the commercial biowaste scenario (manure “ex-housing” + commercial biowaste “ex-hygienisation”).

	Manure ("ex housing")	Biowaste from commercials ^a	Mass balance: amount in manure	Mass balance: amount in biowaste	Biomass mixture entering digester
	kg /	kg /	kg /	kg /	kg /
Unit	1 000.0 kg manure “ex housing”	1 000.0 kg biowaste	370.7 kg manure “ex housing”	629.3 kg biowaste	1 000.0 kg biomass mixture
DM	68.7	244.1	25.5	153.6	179.1
VS	54.6	228.4	20.2	143.7	163.9
Total N	5.26	8.06	1.95	5.07	7.02
Phosphorus (P)	1.21	1.12	0.45	0.70	1.15
Potassium (K)	2.85	2.20	1.06	1.38	2.44
Carbon (C)	34.25	127.19	12.69	80.04	92.74
Copper (Cu)	0.031	0.009	0.011	0.006	0.017
Zinc (Zn)	0.091	0.002	0.034	0.001	0.035

^a See Table S58.

The emissions due to the anaerobic digestion process, as well as the heat and electricity consumed were modelled using the same algorithms as described in Table S9, but the CO₂/CH₄ ratio (for calculating the biogenic CO₂ releases) was here considered as 2.06 (see section 5).

As a result of the biogas production, the composition of the digestate, as it leaves the digester, will be different as the composition of mixture that entered the digester. This is shown in Table S38.

Table S38. Mass balance for the biomass mixture (commercial biowaste scenario) before and after the anaerobic digestion process.

	Biomass mixture entering digester	Mass balance: Change during biogas production	Mass balance: Amount after biogas production	Digestate "ex biogas plant"
Unit	kg/1 000.0 kg biomass mixture	kg	kg	kg/1 000.0 kg digestate
Total mass	1 000.0	- 82.5 ^a	917.5	1000
DM	179.1	- 82.5 ^b	96.6	105.3
VS	163.9	- 82.5 ^c	81.4	88.8
Total N	7.02	No change	7.02	7.65
Phosphorus (P)	1.15	No change	1.15	1.26
Potassium (K)	2.44	No change	2.44	2.66
Carbon (C)	92.74	- 38.75 ^d	53.99	58.84
Copper (Cu)	0.017	No change	0.017	0.019
Zinc (Zn)	0.035	No change	0.035	0.038

^a This loss corresponds to the biogas produced (71.2 Nm³ biogas per t mixture), expressed in mass terms (density of 1.158 kg Nm⁻³);

^b No water loss and therefore change in dry matter is equal to change in total mass;

^c The same change as for DM (all DM lost was VS);

^d This corresponds to the losses in the biogas itself and the losses that occurred during the digestion process; estimated as described in Table S10.

3.4.3 Co-generation of heat and power (commercial biowaste scenario)

This process was modeled as described in section 3.1.6.

3.4.4 Avoided heat production (commercial biowaste scenario)

This process was modeled as described in section 3.1.7.

3.4.5 Avoided electricity production (commercial biowaste scenario)

This process was modeled as described in section 3.1.8.

3.4.6 Storage of the digestate (commercial biowaste scenario)

For this process, the life cycle inventory is based on the same algorithms as shown in Table S14, except that a value of 2.06 is considered for the ratio CO₂: CH₄. The B₀ calculated for the mixture (based on the methane potential of each substrate, and their proportion in the mixture) was 0.486 m³ CH₄/kg VS. Further, it was here considered that 77% of the digestate's N is TAN (considering that 77% of the N is TAN for both fractions).

Table S39 presents the mass balance related to this process.

Table S39. Mass balance of the digestate (commercial biowaste scenario) before and after storage

	Digestate "ex biogas plant"	Mass balance: Change during storage of digestate	Mass balance: Amount after storage of digestate	Digestate "ex storage"
Unit	kg/1000 kg digestate "ex biogas plant"	kg	kg	kg/1000 kg digestate "ex storage"
Total mass	1000	17.5 ^a	1018	1000.0
DM	105.3	- 2.47 ^b	102.8	101.0
VS	88.8	- 2.47 ^c	86.3	84.8
Total N	7.65	- 0.20 ^d	7.46	7.33
Phosphorus (P)	1.26	No change	1.26	1.24
Potassium (K)	2.66	No change	2.66	2.61
Carbon (C)	58.84	- 2.28 ^d	56.57	55.59
Copper (Cu)	0.019	No change	0.019	0.0184
Zinc (Zn)	0.038	No change	0.038	0.0376

^a Same water addition as for the maize scenario, minus DM loss (given below);

^b The change is calculated as a sum of N and C losses; the authors acknowledge that the value is underestimated, but this rough assumption was allowed as the value is irrelevant in the further life cycle stages;

^c Estimated as equal to DM (all DM losses were VS);

^d Estimated as in the maize scenario;

3.4.7 Digestate spreading on land (commercial biowaste scenario)

This process was modeled as described in Table S16.

3.4.8 Avoided biowaste combustion (commercial biowaste scenario)

As described in the main manuscript, if commercial biowaste would not have been used for anaerobic digestion, it is considered that it would have been used for combustion in a municipal solid waste incineration CHP plant, with electricity and heat efficiencies corresponding to 20.7% and 74% respectively, and a LHV of 20.00 MJ kg⁻¹ DM (as for household biowaste). Based on the DM content of the biowaste, this corresponds to a LHV of 4.9 MJ kg⁻¹ biowaste.

The life cycle inventory for this process is exactly as in Table S35, except for the flow of Cu (9.24E-07 kg) and of biogenic CO₂ (4.65E+02 kg). The induced heat and electricity produced due to the energy no longer provided by commercial biowaste combustion is as shown in Figure S4.

3.4.9 Reference manure handling avoided: storage and spreading

These processes are as described in sections 3.1.14 and 3.1.15.

3.5 Garden waste scenario

3.5.1 Shredding of the garden waste

These processes are as described in the main manuscript.

3.5.2 Biogas production (garden waste scenario)

Here, the same considerations as described in section 3.1.5 for the maize silage scenario apply. Based on the VS biodegradability (Table S40), composition and methane yields considered for garden waste (203 Nm³ per t VS) and manure (319 Nm³ per t), the biomass mixture was calculated to consist of 77.15% manure (ex-housing) and 22.85% garden waste.

Table S40. VS biodegradability, garden waste scenario

Digestion step	Manure: % of initial VS degraded	Garden waste: % of initial VS degraded
Step 1	60 ^c	62 ^c
Step 2 (including step 1)	66 ^a	68 ^b

^a Taken according to [1].

^b See Table S58.

^c Corresponds to 90 % of degradability at step 2.

The composition of the mixture entering the digester and mass balances are shown in Table S41.

Table S41. Composition of the biomass mixture for the garden waste scenario (manure “ex-housing” + garden waste).

	Manure ("ex housing")	Garden waste ^a	Mass balance: amount in manure	Mass balance: amount in garden waste	Biomass mixture entering digester
Unit	kg / 1 000.0 kg manure “ex housing”	kg / 1 000.0 kg garden waste	kg / 771.5 kg manure “ex housing”	kg / 228.5 kg garden waste	kg / 1 000.0 kg biomass mixture
DM	68.7	609.0	53.0	139.1	192.1
VS	54.6	517.0	42.1	118.1	160.2
Total N	5.26	3.41	4.06	0.78	4.84
Phosphorus (P)	1.21	0.67	0.94	0.15	1.09
Potassium (K)	2.85	6.09	2.20	1.39	3.59
Carbon (C)	34.25	163.21	26.42	37.29	63.71
Copper (Cu)	0.031	0.039	0.024	0.009	0.033
Zinc (Zn)	0.091	0.008	0.070	0.002	0.072

^a See Table S58.

The emissions due to the anaerobic digestion process, as well as the heat and electricity consumed were modeled using the same algorithms as described in Table S9, but the CO₂/CH₄ ratio (for calculating the biogenic CO₂ releases) was here considered as 1.95 (see section 5).

As a result of the biogas production, the composition of the digestate, as it leaves the digester, will be different as the composition of mixture that entered the digester. This is shown in Table S42.

Table S42. Mass balance for the biomass mixture (garden waste scenario) before and after the anaerobic digestion process.

	Biomass mixture entering digester	Mass balance: Change during biogas production	Mass balance: Amount after biogas production	Digestate "ex biogas plant"
Unit	kg/1 000.0 kg biomass mixture	kg	kg	kg/1 000.0 kg digestate
Total mass	1000.0	-66.6 ^a	933.4	1000
DM	192.1	-66.6 ^b	125.6	134.5
VS	160.2	-66.6 ^c	93.6	100.3
Total N	4.84	No change	4.84	5.18
Phosphorus (P)	1.09	No change	1.09	1.17
Potassium (K)	3.59	No change	3.59	3.85
Carbon (C)	63.71	-31.26 ^d	32.45	34.77
Copper (Cu)	0.033	No change	0.033	0.035
Zinc (Zn)	0.072	No change	0.072	0.077

^a This loss corresponds to the biogas produced (57.5 Nm³ biogas per t mixture), expressed in mass terms (density of 1.158 kg Nm⁻³);

^b No water loss and therefore change in dry matter is equal to change in total mass;

^c The same change as for DM (all DM lost was VS);

^d This corresponds to the losses in the biogas itself and the losses that occurred during the digestion process; estimated as described in Table S10.

3.5.3 Co-generation of heat and power (garden waste scenario)

This process was modeled as described in section 3.1.6.

3.5.4 Avoided heat production (garden waste scenario)

This process was modeled as described in section 3.1.7.

3.5.5 Avoided electricity production (garden waste scenario)

This process was modeled as described in section 3.1.8.

3.5.6 Storage of the digestate (garden waste scenario)

For this process, the life cycle inventory is based on the same algorithms as shown in Table S14, except that a value of 1.95 is considered for the ratio CO₂: CH₄. The B₀ calculated for the mixture (based on the methane potential of each substrate, and their proportion in the mixture) was 0.441 m³ CH₄/kg VS. Further, it was here considered that 77% of the digestate's N is TAN (considering that 77% of the N is TAN for both fractions).

Table S43 presents the mass balance related to this process.

Table S43. Mass balance of the digestate (garden scenario) before and after storage

	Digestate "ex biogas plant"	Mass balance: Change during storage of digestate	Mass balance: Amount after storage of digestate	Digestate "ex storage"
Unit	kg/1000 kg digestate "ex biogas plant"	kg	kg	kg/1000 kg digestate "ex storage"
Total mass	1000	17.6 ^a	1 018	1 000.0
DM	134.5	- 2.41 ^b	132.1	129.8
VS	100.3	- 2.41 ^c	97.9	96.2
Total N	5.18	- 0.13 ^d	5.05	4.96
Phosphorus (P)	1.17	No change	1.17	1.15
Potassium (K)	3.85	No change	3.85	3.78
Carbon (C)	34.77	- 2.28 ^d	32.49	31.93
Copper (Cu)	0.035	No change	0.035	0.035
Zinc (Zn)	0.077	No change	0.077	0.076

^a Same water addition as for the maize scenario, minus DM loss (given below);

^b The change is calculated as a sum of N and C losses; the authors acknowledge that the value is underestimated, but this rough assumption was allowed as the value is irrelevant in the further life cycle stages;

^c Estimated as equal to DM (all DM losses were VS);

^d Estimated as in the maize scenario;

3.5.7 Digestate spreading on land (garden waste scenario)

This process was modeled as described in Table S16.

3.5.8 Avoided composting (garden waste scenario)

As described in the main manuscript, if garden waste would not have been used for anaerobic digestion, it is considered that it would have been composted (open windrow composting). The output of the composting process consists of screened wooden materials with 64% DM, and mature compost with 68% DM (main manuscript). The compost is then stored in a completely covered storage facility and applied on land, while the wood chips are burned in a small-to-medium scale biomass CHP plant with the same efficiencies as for straw.

The inventory data considered for the composting process are presented in Table S44.

Table S44. Life cycle inventory for the (avoided) garden waste composting process

Input		
Garden waste for composting	1 000.0 kg	The process is related to 1 000.0 kg garden waste.
Output		
Compost "mature"	419.8 kg	Compost amount after screening (Table S45)
Wood chips	74.7 kg	Amount of wood chips obtained from screening (Table S45)
Energy consumption		
Diesel	3.00 l	Diesel consumption for windrow composting. Based on [48].
Electricity	0.2 kWh	Electricity consumption for lighting, heating of engines (machinery) and use of electricity in administration buildings. Based on [49].
Emission to air		
Carbon dioxide biogenic (CO ₂)	327.62 kg	Corresponds to the difference between the total C loss and C lost as CH ₄ (given below): $[91.40 \text{ kg C} - 2.73 \text{ kg CH}_4 * (12/16)] * (44/12) = 327.62 \text{ kg CO}_2$.
Methane (CH ₄)	2.73 kg	Based on [48], corresponds to the 2.24 % of the total C loss: $163.21 \text{ kg C (Table S45)} * 2.24 \% * (16/12) = 2.73 \text{ kg CH}_4$ produced.
Nitrous oxide (N ₂ O-N)	0.04 kg	Based on [48], corresponds to the 15 % of the total N loss: $0.27 \text{ kg N (Table S45)} * 15 \% = 0.04 \text{ kg N}_2\text{O-N}$.
Ammonia (NH ₃ -N)	0.23 kg	Based on [48], corresponds to the 83 % of the total N loss: $0.27 * 83 \% = 0.23 \text{ kg NH}_3\text{-N}$.
Indirect N ₂ O-N (volatilization)	0.0023 kg	Indirect emissions due to volatilization of ammonia (NH ₃ -N) and nitrogen oxides (NO _x -N). Based on IPCC guidelines, correspond to 0.01 kg N ₂ O-N per kg (NH ₃ -N + NO _x -N) volatilized
Carbon monoxide (CO)	0.725 kg	Based on [50], CO-C corresponds to 0.34% of the C losses.
Discharge to water		
	None	No losses are considered.
Discharge to soil		
	None	No losses are considered.

The mass balances related to the composting (and screening of the woody material) process is shown in Table S45.

Table S45. Mass balance for garden waste composting

	Garden waste ^a	Mass balance: Change during composting	Mass balance: Amount after composting	Mass balance: Amount in compost "mature"	Mass balance: Amount in wood chips	Compost "mature"	Wood chips
Unit	kg/ 1 000.0 kg biomass initial	kg	kg	kg	kg	kg/ 1 000.0 kg compost "mature"	kg/ 1 000.0 kg wood chips
Total mass	1000.0	-505.5 ^b	494.5	419.8 ^l	74.7 ^m	1000.0	1000.0
DM	609.0	-289.5 ^c	319.5	268.7 ^k	50.8 ^k	640.0 ⁱ	680.0 ^j
VS	517.0	-289.5 ^d	227.5	191.3 ^g	36.2 ^h	455.7	484.2
Total N	3.41	-0.27 ^e	3.14	2.64 ^k	0.50 ^k	6.29	6.68
Phosphorus (P)	0.67	No change	0.67	0.56 ^k	0.11 ^k	1.34	1.43
Potassium (K)	6.09	No change	6.09	5.12 ^k	0.97 ^k	12.20	12.96
Carbon (C)	163.21	-91.40 ^f	71.81	60.39 ^k	11.42 ^k	143.86	152.85
Copper (Cu)	0.039	No change	0.03928	0.033 ^k	0.006 ^k	0.079	0.084
Zinc (Zn)	0.008	No change	0.00792	0.007 ^k	0.001 ^k	0.016	0.017

^a All the data are the same as given for garden waste in Table S58;

^b Taken as a difference between initial mass (1 000.0 kg garden waste) and mass of output products (419.9 kg compost "mature" and 74.7 kg wood chips; established as described in "i" and "j" below);

^c Based on [49], no ash losses is expected, thus, the same change as for VS is assumed;

^d Based on [49], corresponds to 56 % of the initial VS;

^e Based on [48], corresponds to 8 % of the initial N;

^f Based on [49], corresponds to 56% of the initial C;

^g Based on [49], correspond to 37 % of the initial VS;

^h Based on [49], corresponds to 7 % of the initial VS;

ⁱ Based on [48], set at 64 % of the total weight;

^j Based on [48], set at 68 % of the total weight.

^k Calculated proportional to VS allocation;

^l Calculated from DM amount in "compost mature" (backwards calculation)

^m Calculated from DM amount in "wood chips" (backwards calculation)

3.5.9 Storage of the compost (avoided) (garden waste scenario)

As earlier described, the compost is assumed to be stored in a facility completely protected from rain addition. The life cycle inventory considered for this process is presented in Table S46.

Table S46. Life cycle inventory for process “avoided compost storage”, garden waste scenario

Comments		
Input		
Compost "mature"	1 000.0 kg	The process is related to 1 000.0 kg compost "mature". The emissions are calculated relative to this.
Water	0	Assumed to be none.
Output		
Compost ("ex storage")	998.9 kg	The compost amount after storage considering total C and N losses.
Emission to air		
Carbon dioxide (CO ₂)	1.066 kg	Calculated from CH ₄ emissions using the ratio kg CO ₂ /kg CH ₄ for garden waste (2.33) as established in section 5.
Methane (CH ₄)	0.457 kg	Calculated with IPCC guidelines using MCF = 0.5 % and B ₀ = 0.299 m ³ CH ₄ /kg VS (methane potential of garden waste): 455.7 kg VS/t compost * 0.299 m ³ CH ₄ /kg VS * 0.67 kg CH ₄ /m ³ CH ₄ * 0.5 % = 0.457 kg.
Ammonia (NH ₃ -N)	0.314 kg	Calculated using an emission factor of 5 % of total N in compost ending up as NH ₃ -N, based on [11], for solid cow manure storage.
Direct emissions of Nitrous oxide (N ₂ O-N)	0.0314 kg	Calculated using an emission factor of 0.005 total N in compost ending up as NH ₃ -N, based on [12].
Indirect emissions of Nitrous oxide (N ₂ O-N)	0.00346 kg	Indirect emissions due to volatilization of ammonia (NH ₃ -N) and nitrogen oxides (NO _x -N). Based on IPCC guidelines, correspond to 0.01 kg N ₂ O-N per kg (NH ₃ -N + NO _x -N) volatilized.
Nitrogen monoxide (NO-N) (representing total NO _x)	0.0314 kg	Based on [51], estimated as: NO-N = (direct) N ₂ O-N * 1.
Nitrogen (N ₂ -N)	0.0943 kg	Based on [51], estimated as: N ₂ -N = (direct) N ₂ O-N * 3.
Discharge to water		
	None	Assumed to be none.

Table S47 presents the mass balance of the compost as it enters and leaves the storage.

Table S47. Mass balance for the garden waste compost before and after storage

	Compost "mature"	Mass balance: Change during storage	Mass balance: Amount after storage	Compost "ex storage"
Unit	kg/1 000.0 kg compost "mature"	kg	kg	kg/1 000.0 kg compost "ex storage"
Total mass	1 000.0	-1.1 ^a	998.9	1000.0
DM	640.0	-1.1 ^b	638.9	639.6
VS	455.7	-1.1 ^c	454.6	455.1
Total N	6.3	-0.47 ^d	5.81	5.82
Phosphorus (P)	1.3	No change	1.34	1.34
Potassium (K)	12.2	No change	12.20	12.21
Carbon (C)	143.9	-0.63 ^d	143.23	143.39
Copper (Cu)	0.1	No change	0.079	0.079
Zinc (Zn)	0.016	No change	0.016	0.016

^a Equal to the DM loss (given below);

^b The change is calculated as a sum of N and C losses; the authors acknowledge that the value is underestimated, but this rough assumption was allowed as the value is irrelevant in the further life cycle stages;

^c Estimated as equal to DM (all DM losses were VS);

^d Estimated as in the maize scenario;

3.5.10 Avoided compost application on land (garden waste scenario)

This process was modeled as described in Table S16, but with the 3 following changes. Firstly, a diesel consumption of 12 L ha⁻¹ was considered for the application of the compost, based on [49]. Second, no "NH₃ emissions during the very moment of application" were considered, as incorporation was assumed as the application technique. Thirdly, the NH₃-N emissions "in the period after application" were estimated as 9.25% of the total N applied, based on [11], for solid manure application.

3.5.11 Avoided wood chips combustion

The screened wood chips from the composting process would – if the composting process had occurred – have been burned in a small-to-medium scale biomass CHP plant with efficiencies assumed to 27% and 63% for electricity and heat, respectively (exactly as for straw combustion). The LHV used for wood chips is 18.100 MJ/kg wood chip DM [30], which here corresponds to 12.308 MJ/kg wood chip wet weight.

The life cycle inventory for this process is presented in Table S48.

Table S48. Life cycle data for wood chips combustion in CHP plant (garden waste scenario)

Comments		
Input		
Wood chips	0.081 kg	Amount of wood chips corresponding to an energy content of 1 MJ input: $1 \text{ MJ}/12.308 \text{ MJ/kg wood chips} = 0.081 \text{ kg wood chips}$
Output		
Heat	0.63 MJ	The efficiency of the heat production is 63 %, see text.
Electricity	0.27 MJ	The electricity efficiency is 27 %, see text.
Emission to air		
Carbon dioxide (CO ₂), biogenic	4.54 E-02 kg	Estimated as the difference between the total C in the 0.081 kg wood chips, and the CH ₄ -C and CO-C losses (given below): $(0.081 \text{ kg wood chips} * 152.85 \text{ kg C}/1000.0 \text{ kg wood chips} - (3.10 * 10^{-6} * 12/16 + 9.00 * 10^{-5} * 12/28)) * 44/12 = 0.0454 \text{ kg CO}_2$.
Carbon dioxide (CO ₂), fossil	5.15 E-03 kg	Due to the fossil fuel consumption needed in order to run the process. Taken as for biowaste incineration, but adjusted to be expressed per wood chip quantity, i.e.: $0.081 \text{ kg wood chips} * 63.4 \text{ kg CO}_2 / 1000.0 \text{ kg} = 5.15 * 10^{-3} \text{ kg CO}_2$.
Sulphur dioxide (SO ₂)	1.90 E-06 kg	Reference [25]
Nitrogen oxides (NO _x)	8.10 E-05 kg	Reference [25]
Non-methane volatile organic compounds (NMVOC)	5.10 E-06 kg	Reference [25]
Methane (CH ₄)	3.10 E-06 kg	Reference [25]
Carbon monoxide (CO)	9.00 E-05 kg	Reference [25]
Nitrous oxide (N ₂ O)	8.00 E-07 kg	Reference [25]
TSP	1.00 E-05 kg	Reference [25]
Cd	2.70 E-10 kg	Reference [25]
Hg	4.00 E-10 kg	Reference [25]
Zn	2.30 E-09 kg	Reference [25]
PCDD/-F ^o	1.40 E-14 kg	Reference [25]

^o Modelled as “Dibenzofuran”.

The induced heat and electricity produced due to the energy no longer provided by the wood chips combustion is as shown in Figure S5.

3.5.12 Reference manure handling avoided: storage and spreading

These processes are as described in sections 3.1.14 and 3.1.15.

3.6 Manure source-segregation scenario

3.6.1 Source-segregation of manure

As described in the main manuscript, this scenario considers that the raw pig slurry is co-digested with a concentrated solid fraction obtained from source-segregation of urine and feces in the animal house (of a second farm). The separation technology considered is based on the rotating belt conveyor technology developed by Lemay and coll. [52–54], and the separation efficiencies considered for this technology are presented in Table S49.

Table S49. Separation efficiencies considered for the source-segregation of manure

	Segregation efficiency ^a
Total mass	16%
DM	72%
VS	77%
Total N	42%
Phosphorus (P)	79%
Potassium (K)	28%
Carbon (C)	77%
Copper (Cu)	82%
Zinc (Zn)	76%

^a These % indicate the share transferred to the solid fraction.

Based on this, the composition of the segregated liquid and solid fractions can be calculated (Table S50).

Table S50. Mass balance for the manure ex-animal before and after segregation

	Manure ("ex- animal")	Cleaning water	Mass balance: amount in manure solid fraction ^b	Mass balance: amount in manure liquid fraction ^b	Manure solid fraction "ex segregation"	Manure liquid fraction "ex segregation"
Unit	kg/1 000.0 kg manure "ex- animal"	kg/1 000.0 kg manure "ex- animal"	kg	kg	kg/1 000.0 kg solid fraction "ex- segregation"	kg/1 000.0 kg liquid fraction "ex- segregation"
Total mass	1000.0	237 ^a	195.4	1041.6	1000.0	1000.0
DM	74.8	-	53.8	21.0	275.2	20.2
VS	60.7	-	46.8	13.9	239.2	13.4
Total N	6.00	-	2.5	3.48	12.88	3.34
Phosphorus (P)	1.21	-	0.96	0.25	4.90	0.24
Potassium (K)	2.83	-	0.79	2.04	4.05	1.96
Carbon (C)	34.46	-	26.6	7.90	135.90	7.59
Copper (Cu)	0.031	-	0.03	0.006	0.130	0.005
Zinc (Zn)	0.091	-	0.07	0.022	0.351	0.021

^a As the conveyor belt is rotating, a certain amount of water is used to clean it continuously, so it is clean when it comes in contact with the pigs. The estimation presented here is based on [54], and corresponds to the difference in total volume between the slats

and the conveyor belt treatment, for all treatments (except those with a water pipeline problem). This cleaning water is assumed to be entirely transferred to the liquid fraction.

^b Estimated considering the segregation efficiencies presented in Table S49.

The electricity consumed for running the conveyor is not specified in [54]. Therefore, a rough approximation of 1.2 kWh per t of slurry ex-animal has been assumed, based on the energy needed for stirring slurry [8].

3.6.2 In-house storage of the segregated liquid and solid fraction

A very temporal storage (less than 1 week) of the segregated fractions in-house is considered. The life cycle inventory process related to the storage of these fractions in-house is presented in Table S51. Only the values from the last column (weighted sum from both fractions) were used in the model.

Table S51. Life cycle inventory data for the storage of the liquid and solid manure segregated fractions in-house.

Input	Solid fraction "ex segregation"	Comments	Liquid fraction "ex segregation"	Comments	Manure "ex animal"	Comments
Manure fraction "ex segregation"	1000.0 kg	Related to 1 000.0 kg of manure solid fraction "ex segregation".	1000.0 kg	Related to 1 000.0 kg of manure liquid fraction "ex segregation".	1000.0 kg	Related to 1 000 kg manure "ex animal" ^a .
Output						
Manure fraction "ex housing"	996.7 kg	Manure solid fraction leaving the storage.	996.9 kg	Manure liquid fraction leaving the storage.		
Emission to air						
Carbon dioxide (CO ₂)	3.24 kg	Same algorithm as in Table S2, for in-house storage.	0.18 kg	Same algorithm as in Table S2, for in-house storage.	0.824 kg	Calculated as the weighted sum for each fraction.
Methane (CH ₄)	1.77 kg	Calculated using the algorithm of [17], for a storage duration of 7 days, and a temperature of 17.5°C.	0.10 kg	Calculated using the algorithm of [17], for a storage duration of 7 days, and a temperature of 17.5°C.	0.450 kg	Calculated as the weighted sum for each fraction.
Ammonia (NH ₃ -N)	0.32 kg	Same algorithm as in Table S2, for in-house storage, but an emission reduction factor of 14% is applied, based on the results of [54].	0.30 kg	Same algorithm as in Table S2, for in-house storage, but an emission reduction factor of 17% is applied, based on the results of [54].	0.378 kg	Calculated as the weighted sum for each fraction.
Direct emissions of Nitrous oxide (N ₂ O-N)	0.064 kg	Same algorithm as in Table S2, for in-house storage, but using the IPCC factor for solid storage (0.005).	0.007 kg	Same algorithm as in Table S2, for in-house storage.	0.020 kg	Calculated as the weighted sum for each fraction.
Indirect emissions of Nitrous oxide (N ₂ O-N)	0.003 kg	Same algorithm as in Table S2, for in-house storage.	0.003 kg	Same algorithm as in Table S2, for in-house storage.	0.00381 kg	Calculated as the weighted sum for each fraction.
Nitrogen monoxide (NO-N) (representing total NO _x)	0.0104 kg	Based on [7], corresponds to 1% of the TAN (for solid manure).	0.0001 kg	Same algorithm as in Table S2, for in-house storage.	0.00213 kg	Calculated as the weighted sum for each fraction.
Nitrogen (N ₂ -N)	0.67 kg	Based on [7], corresponds to 30% of the TAN (for solid manure).	0.006 kg	Same algorithm as in Table S2, for in-house storage.	0.137 kg	Calculated as the weighted sum for each fraction.
Discharge to water and soil						
	None		None		Assumed to be zero.	

^a Corresponding to handling 195.4 kg manure solid fraction "ex segregation" and 1041.6 kg manure liquid fraction "ex segregation". The values in this column were those used to model this process.

The calculation of the composition of each fraction before and after storage is presented in Table S52-Table S53.

Table S52. Mass balance for the solid fraction before and after in-house storage

	Manure solid fraction "ex segregation"	Mass balance: Change during storage	Mass balance: Amount after storage	Manure solid fraction "ex housing"
Unit	kg/1000 kg manure solid fraction "ex segregation"	kg	kg	kg/1000 kg manure solid fraction "ex housing"
Total mass	1000.0	-3.3 ^a	996.7	1000.0
DM	275.2	-3.3 ^a	271.9	272.8
VS	239.2	-3.3 ^b	236.0	236.7
Total N	12.88	-1.07 ^c	11.81	11.85
Phosphorus (P)	4.90	No change	4.90	4.92
Potassium (K)	4.05	No change	4.05	4.06
Carbon (C)	135.90	-2.21 ^d	133.69	134.13
Copper (Cu)	0.130	No change	0.130	0.130
Zinc (Zn)	0.351	No change	0.351	0.352

^a The change in DM and in total mass is assumed to be identical to the sum of N and C losses;

^b Assumed equal to DM (i.e. all DM lost was VS).

^c Changes in total N equal to the sum of N losses due to N-emissions occurring during the storage.

^d Changes in total C equal to the sum of C losses due to CO₂ and CH₄ emissions occurring during the storage.

Table S53. Mass balance for the liquid fraction before and after storage

	Manure liquid fraction "ex segregation"	Mass balance: Change during storage	Mass balance: Amount after storage	Manure liquid fraction "ex housing"
Unit	kg/1000 kg manure liquid fraction "ex segregation"	kg	kg	kg/1000 kg manure liquid fraction "ex housing"
Total mass	1000.0	-0.4 ^a	999.6	1000.0
DM	20.2	-0.4 ^a	19.8	19.8
VS	13.4	-0.4 ^b	12.9	12.9
Total N	3.34	-0.32 ^c	3.03	3.03
Phosphorus (P)	0.24	No change	0.24	0.24
Potassium (K)	1.96	No change	1.96	1.96
Carbon (C)	7.59	-0.12 ^d	7.46	7.47
Copper (Cu)	0.005	No change	0.005	0.005
Zinc (Zn)	0.021	No change	0.021	0.021

^a The change in DM and in total mass is assumed to be identical to the sum of N and C losses;

^b Assumed equal to DM (i.e. all DM lost was VS).

^c Changes in total N equal to the sum of N losses due to N-emissions occurring during the storage.

^d Changes in total C equal to the sum of C losses due to CO₂ and CH₄ emissions occurring during the storage.

3.6.3 Biogas production (manure segregation scenario)

Here, the same considerations as described in section 3.1.5 for the maize silage scenario apply. The methane yield and VS degradability considered for the segregated solid fraction are the same as for raw manure, based on [1]. Based on this, the mixture consists of 41.02% raw manure, and 58.98% segregated solid manure.

The composition of the mixture entering the digester and mass balances are shown in Table S54.

Table S54. Composition of the biomass mixture for the segregated manure scenario (manure “ex-housing” + segregated solid fraction “ex-housing”).

	Manure ("ex housing")	Manure solid fraction (ex- housing)	Mass balance: amount in manure	Mass balance: amount in manure solid fraction	Biomass mixture entering digester
Unit	kg / 1 000.0 kg manure “ex housing”	kg / 1 000.0 kg manure solid fraction “ex housing”	kg / 410.2 kg manure “ex-housing”	kg / 589.8 kg segregated solid manure “ex- housing”	kg / 1 000.0 kg biomass mixture
DM	68.7	272.8	28.2	160.9	189.1
VS	54.6	236.7	22.4	139.6	162.0
Total N	5.26	11.85	2.16	6.99	9.15
Phosphorus (P)	1.21	4.92	0.50	2.90	3.40
Potassium (K)	2.85	4.06	1.17	2.40	3.57
Carbon (C)	34.25	134.13	14.05	79.11	93.16
Copper (Cu)	0.031	0.130	0.013	0.077	0.089
Zinc (Zn)	0.091	0.352	0.037	0.208	0.245

The emissions due to the anaerobic digestion process, as well as the heat and electricity consumed were modeled using the same algorithms as described in Table S9, but the CO₂/CH₄ ratio (for calculating the biogenic CO₂ releases) was here considered as 1.83 (see section 5).

As a result of the biogas production, the composition of the digestate, as it leaves the digester, will be different as the composition of mixture that entered the digester. This is shown in Table S55.

Table S55. Mass balance for the biomass mixture (manure segregation scenario) before and after the anaerobic digestion process.

	Biomass mixture entering digester	Mass balance: Change during biogas production	Mass balance: Amount after biogas production	Digestate "ex biogas plant"
Unit	kg/1 000.0 kg biomass mixture	kg	kg	kg/1 000.0 kg digestate
Total mass	1 000.0	- 92.0 ^a	908.0	1 000.0
DM	189.1	- 92.0 ^b	97.1	106.9
VS	162.0	- 92.0 ^c	70.0	77.1
Total N	9.15	No change	9.15	10.07
Phosphorus (P)	3.40	No change	3.40	3.74
Potassium (K)	3.57	No change	3.57	3.93
Carbon (C)	93.16	- 43.20 ^d	49.96	55.03
Copper (Cu)	0.089	No change	0.089	0.098
Zinc (Zn)	0.245	No change	0.245	0.270

^a This loss corresponds to the biogas produced (79.5 Nm³ biogas per t mixture), expressed in mass terms (density of 1.158 kg Nm⁻³);

^b No water loss and therefore change in dry matter is equal to change in total mass;

^c The same change as for DM (all DM lost was VS);

^d This corresponds to the losses in the biogas itself and the losses that occurred during the digestion process; estimated as described in Table S10.

3.6.4 Co-generation of heat and power (manure segregation scenario)

This process was modeled as described in section 3.1.6.

3.6.5 Avoided heat production (manure segregation scenario)

This process was modeled as described in section 3.1.7.

3.6.6 Avoided electricity production (manure segregation scenario)

This process was modeled as described in section 3.1.8.

3.6.7 Storage of the digestate (manure segregation scenario)

For this process, the life cycle inventory is based on the same algorithms as shown in Table S14, except that a value of 1.83 is considered for the ratio CO₂: CH₄. The B₀ calculated for the mixture (based on the methane potential of each substrate, and their proportion in the mixture) was 0.483 m³ CH₄/kg VS. Further, it was here considered that 43% of the digestate's N is TAN (considering that 77% of the N is TAN for the raw manure portion, and 19% for the segregated manure solid fraction portion).

Table S56 presents the mass balance related to this process.

Table S56. Mass balance of the digestate (manure segregation scenario) before and after storage

	Digestate "ex biogas plant"	Mass balance: Change during storage of digestate	Mass balance: Amount after storage of digestate	Digestate "ex storage"
Unit	kg/1000 kg digestate "ex biogas plant"	kg	kg	kg/1000 kg digestate "ex storage"
Total mass	1 000.0	18.0 ^a	1 017.9	1000
DM	106.9	- 2.04 ^b	104.9	103.0
VS	77.1	- 2.04 ^c	75.1	73.7
Total N	10.07	- 0.17 ^d	9.91	9.73
Phosphorus (P)	3.74	No change	3.74	3.68
Potassium (K)	3.93	No change	3.93	3.86
Carbon (C)	55.03	- 1.87 ^d	53.16	52.22
Copper (Cu)	0.098	No change	0.098	0.097
Zinc (Zn)	0.270	No change	0.270	0.265

^a Same water addition as for the maize scenario, minus DM loss (given below);

^b The change is calculated as a sum of N and C losses; the authors acknowledge that the value is underestimated, but this rough assumption was allowed as the value is irrelevant in the further life cycle stages;

^c Estimated as equal to DM (all DM losses were VS);

^d Estimated as in the maize scenario;

3.6.8 Digestate spreading on land (manure segregation scenario)

This process was modeled as described in Table S16.

3.6.9 Storage of the liquid separated fraction (manure segregation scenario)

For this process, the life cycle inventory is based on the same algorithms as shown in Table S14, except that a value of 1.83 is considered for the ratio CO₂: CH₄. The B₀ calculated for the liquid fraction was 0.483 m³ CH₄/kg VS (as for raw manure). Further, it was here considered that 62% of the digestate's N is TAN (based on the measurement results shown in [54]).

Table S57 presents the mass balance related to this process.

Table S57. Mass balance of the liquid fraction before and after outdoor storage.

	Manure liquid fraction "ex housing"	Mass balance: Change during storage	Mass balance: Amount after storage	Manure liquid fraction "ex storage"
Unit	kg/1000 kg manure liquid fraction "ex housing"	kg	kg	kg/1000 kg manure liquid fraction "ex storage"
Total mass	1000	19.4 ^a	1019.7	1000.0
DM	19.8	-0.6 ^b	19.2	18.8
VS	12.9	-0.6 ^c	12.3	12.1
Total N	3.03	-0.07 ^d	2.96	2.91
Phosphorus (P)	0.24	No change	0.24	0.24
Potassium (K)	1.96	No change	1.96	1.92
Carbon (C)	7.47	-0.52 ^d	6.94	6.81
Copper (Cu)	0.005	No change	0.005	0.005
Zinc (Zn)	0.021	No change	0.021	0.021

^a Same water addition as for the maize scenario, minus DM loss (given below);

^b The change is calculated as a sum of N and C losses; the authors acknowledge that the value is underestimated, but this rough assumption was allowed as the value is irrelevant in the further life cycle stages;

^c Estimated as equal to DM (all DM losses were VS);

^d Estimated as in the maize scenario;

3.6.10 Liquid spreading on land (manure segregation scenario)

This process was modeled as described in Table S16.

3.6.11 Avoided reference manure management at farm 2 (manure segregation scenario)

At the second farm, where manure is source-segregated, it considered that the lost alternative is the reference manure management. In other words, if the manure would not have been separated, it would have been managed according to the conventional manure management, without any treatment. The processes considered for the reference in-house storage, outdoor storage and field application are as described in sections 3.1.1, 3.1.14 and 3.1.15.

3.7 Mono-digestion scenario

This scenario is modeled exactly as previous scenarios (e.g. maize), but with only one tonne of raw slurry (ex-housing) as input to the digester. The lost alternative for this manure is the conventional manure management (storage and application on land).

4. Biowaste potential and composition

The details of the biowaste compositions considered, and on how these were derived, are presented in Table S58.

Table S58. Characteristics of the biowastes considered in this study

	Unit	1) Biowaste from households	2) Garden waste from households and city gardens	3) Biowaste from whole sale and retail commercials (commercial biowaste) ^h
DM		315.0 ^a	609.0 ^e	244.1 ⁱ
VS		259.8 ^b	517.0 ^e	228.4 ⁱ
Total N		8.79 ^a	3.41 ^e	8.06 ⁱ
Phosphorus (P)	kg / 1 000.0 kg wet weight	1.29 ^a	0.67 ^e	1.12 ⁱ
Potassium (K)		2.80 ^a	6.09 ^e	2.20 ^j
Carbon (C)		141.75 ^a	163.21 ^e	127.19 ^j
Copper (Cu)		0.009 ^a	0.008 ^e	0.002 ^j
Zinc (Zn)		0.022 ^a	0.039 ^e	0.009 ^j
Methane yield	Nm ³ CH ₄ / t VS	330 ^c	203 ^f	277 ^k
Degradability in anaerobic digestion	% VS	64 % ^d	68 % ^g	57 % ^l

1)

^a Average based on [55,56].

^b VS = 82 % of TS. This is an average based on [55–59].

^c Average based on [55,56,59].

^d Calculated as methane yield (from above) divided with the methane potential (Table S61).

2)

^e Refers to garden waste at the composting plant in Aarhus. According to [60], the composition of this is constituted of 75.6 % easily degradable material (leaves, grass etc.), 19.5 % branches, 4.5 % woody parts and 0.4 % foreign items.

^f Average of 1) methane yield reported by [57] for garden waste in Kerteminde municipality, and 2) methane yield estimated for garden waste at the Aarhus composting plant. In this latter case, the composition of [60] (described in e) was considered. The methane yield was calculated considering a methane yield of 287 Nm³ CH₄/t VS for grass (average from [20,34,59]) and 160 Nm³ CH₄/t VS for branches and woody parts [61]. Grass is here taken as a representative for the “easily degradable material” portion of the garden waste.

^g Estimated as weighted average (based on the composition of [60], described in e), assuming degradability of 75 % for grass [20,34] and 46 % for branches and woody parts [61].

3)

^h Commercial biowaste is considered to be constituted of whole sale biowaste (24 % fruits and vegetables, 3 % meat products) and retail biowaste (50 % waste from supermarkets and 23 % waste from other food shops).

ⁱ Calculated as a weighted average considering the composition of the biowaste described in h. The composition details from [48] were used for fruits & vegetables as well as meat waste and the composition found in [62] was used for biowaste from supermarkets and other shops (retail biowaste).

^j Average for biowaste from wholesale commercials only (no data for retail commercial biowaste).

^k Calculated as a weighted average considering the composition of the biowaste described in h. Considering methane yields of 450 Nm³ CH₄/t VS for fruits and vegetables and meat waste [48] and 285 Nm³ CH₄/t VS for biowaste from supermarkets and other shops [62].

^l Calculated as methane yield (from above) divided with the methane potential (Table S61).

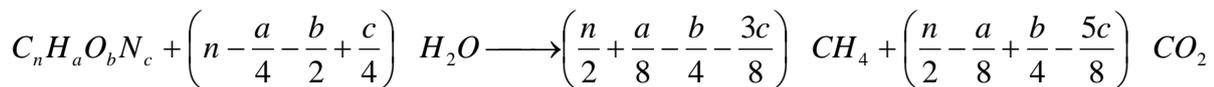
The potential of each biowaste considered, and specifications on how it is used today in Denmark, are presented in Table S59.

Table S59. Biowaste potentials and actual uses in Denmark.

	Total volume, t (wet weight)/ year	Specification on treatment performed today	Reference
1) Biowaste from households	786 616	723 687 Biowaste amount incinerated today, corresponds to 92 % of the total quantity of biowaste from households.	[63]
		54 488 Centralized and home composting.	
		4 628 Amount already used for biogas.	
2) Garden waste from households	700 000	523 000 Garden waste amount composted today.	[63]
		177 000 Garden waste amount estimated to stay inside households.	
Garden waste from city gardens	548 264	246 535 Amount composted today.	[63]
		1 729 Amount incinerated today.	
		300 000 Amounts estimated to stay not collected.	
3) Biowaste from whole sale and retail commercials	93 685	27 756 Amount estimated to derive from whole sale (see note h in Table S58). Assumed to be incinerated.	[63]
		65 929 Amount estimated to derive from retail commercials (see note h in Table S58). Assumed to be incinerated.	

5. CO₂: CH₄ ratio and calculation of methane potential

An original methodology is presented in [1] in order to estimate the ratio between the biogenic CO₂ and CH₄ emitted during anaerobic degradation, and this methodology is applied here. In fact, whenever biogenic CH₄ is emitted (under anaerobic conditions), biogenic CO₂ is simultaneously emitted, as described by the Buswell equation [64]:



(Equation S3)

The first step of the methodology described by [1] consists to determine the relative contribution (in %) of all organic components constituting the VS in the substrate of interest. This was performed in Table S60.

Table S60. Relative contribution of the organic components constituting the VS for each substrate

	Formula	Pig slurry ^a	Maize silage	Straw ^d	HW ^e	CW ^f	GW ^g
VS protein	C ₅ H ₇ O ₂ N	27.0%	6.90 % ^b	4.17 %	18.37%	13.89%	22.22%
VS lipid	C ₅₇ H ₁₀₄ O ₆	16.2%	2.30 % ^b	2.56 %	26.32%	12.73%	4.33%
VS VFA ^h	C ₂ H ₄ O ₂	8.5%	0.00 %	0.00 %	0.00%	0.00%	0.00%
VS carbohydrates (slowly degradable)	C ₆ H ₁₀ O ₅	27.1%	20.20 % ^c	38.85 %	16.42%	0.00%	0.00%
VS carbohydrates (easily degradable)	C ₆ H ₁₀ O ₅	21.1%	70.60 % ^c	54.42 %	38.89%	73.38%	73.44%

^a Based on [65]

^b Based on [21] (variety Tixxus at "wax" ripeness, as it matches best the VS content considered in this study)

^c Calculated with formula of [19], with the data of [21]

^d Based on [19]

^e HW: Household biowaste. Data based on an average from [55,58]

^f CW: Commercial biowaste. Data based on [62]

^g GW: Garden waste. Data based on [9] for permanent meadow grass. It is acknowledged that this is a rough approximation.

^h Except for manure, no data were found on VFA, so these were assumed to be zero. However, since VFA produce the same amount of moles of CO₂ and CH₄ (Table S61), this does not affect the ratio to be calculated.

Information may not be always available to fill in the data required in Table S60. However, the most important in the perspective of deriving a ratio between CO₂ and CH₄ is information on protein and VS, since the amount of moles of CO₂ and CH₄ produced from these is not equal (as this is the case for VFA and carbohydrates). Table S61 shows the ratio between CO₂ and CH₄ for all substrates. The results for slurry differs slightly to those presented by [1], as that study considered a version of the Buswell formula not accounting for the N portion of the organic component (the term N_c). As seen in that Table, the VS slowly degradable are not considered, as these are assumed not to degrade.

Table S61. Calculation of the CO₂: CH₄ ratio from the anaerobic degradation of the substrates considered in this study

	Molecular weight (g/mol)	Moles produced (Buswell equation)		Moles produced, weighted for pig slurry ^a		Moles produced, weighted for maize silage		Moles produced, weighted for straw		Moles produced, weighted for HW ^d		Moles produced, weighted for CW ^d		Moles produced, weighted for garden waste	
		CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂
Protein	113	2.5	1.5	0.006	0.004	0.002	0.001	0.0009	0.0006	0.004	0.002	0.003	0.002	0.005	0.003
Lipid	884	40	17	0.007	0.003	0.001	0.0004	0.0012	0.0005	0.012	0.005	0.006	0.002	0.002	0.0008
VFA	60	1	1	0.001	0.001	0	0	0	0	0	0	0	0	0	0
Carbohydrates (easily degradable)	162	3	3	0.005	0.005	0.013	0.013	0.010	0.010	0.007	0.007	0.014	0.014	0.014	0.014
TOTAL				0.020	0.013	0.016	0.014	0.012	0.011	0.023	0.015	0.022	0.018	0.020	0.017
g CO ₂ /g CH ₄ ^b				1.83		2.54		2.51		1.74		2.19		2.33	
Calculated CH ₄ potential ^c				-	-	-	-	-	-	522		499		-	-

^a Calculation example for protein CH₄: 2.5 moles CH₄/moles protein * 27% (Table S60) / 113 g protein per mole protein = 0.006 moles CH₄

^b Calculation example for slurry: 0.013 moles CO₂/0.020 moles CH₄ * 44 g CO₂/mol CO₂ / 16 g CH₄ per mol CH₄ = 1.83 g CO₂ per g CH₄.

^c Considering a volume of 0.02271 Nm³ CH₄ per mole at normal conditions (0°C, 100 kPa).

^d HW: Household biowaste; CW: Commercial biowaste.

Based on Table S61 and on the share between manure and co-substrate in the mixture input of all scenarios (summarized in Table 1 of the main manuscript), the ratio CO₂:CH₄ can be calculated for each input mixture. The result is shown in Table S62.

Table S62. Ratio CO₂: CH₄ for the input mixtures (to digester) of all scenarios

Scenario	Maize silage	Straw	Household biowaste	Commercial biowaste	Garden waste	Manure segregation	Mono-digestion
g CO ₂ : g CH ₄ of input mixture	2.23	1.94	1.81	2.06	1.95	1.83	1.83

6. Fertilizer substitution

As described in the main manuscript, it was considered that the digestates (and liquid fraction, avoided raw manure or garden waste compost) were applied to the 6-year crop rotation described in [1], for a representative Danish pig farm. The N, P and K requirements of this crop rotation are presented in Table S63.

Table S63. N, P and K requirements of the 6-year crop rotation on which the digestates are applied.

Year	Crop	N (kg ha ⁻¹) ^a	P (kg ha ⁻¹) ^a	K (kg ha ⁻¹) ^a
1	Winter barley	158	21	54
2	Winter rape	177	30	89
3	Winter wheat	166	22	66
4	Winter wheat	166	22	66
5	Spring barley & catch crop	109	22	45
6	Spring barley	126	22	45
Annual average		150	23	61

^a Data for N, P and K requirements are from [66]. It is further considered that catch crops reduce the N norm by 17 kg N ha⁻¹, based on [16].

The calculation of the amount of mineral fertilizers substituted from using the digestates as organic fertilizers was based on the Danish law [67]. 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 not all the N applied with the digestates will end up in an inorganic (or mineralized) form, which can be used by plants, and this is considered in the Danish law (so it can be said that in this sense, a bit of N is applied in excess). This study considers the efficiencies found in the Danish law [67] (Table S64).

As opposed to N, the P and K that can be applied to the field are not limited by law. As a result, they may be applied in excess, since organic fertilizers containing all 3 nutrients (N, P, and K) are applied on the basis of the N requirements only. 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 [1], 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 S63), and the content in P and K in the digestates applied (Table S64).

As a result, the amount of N, P and K avoided is presented in Table S64 (and is illustrated in Figure S1-Figure S7). As mentioned in the manuscript, the mineral fertilizers avoided are considered to be calcium ammonium nitrate, diammonium phosphate and potassium chloride (marginal fertilizers).

Table S64. Calculation of the amount of mineral N, P, and K substituted

"Material" applied on land ^a	Nutrients content in the material applied (kg/t material) ^c			Amount of land needed to apply material (ha/t material) (B) = (C1)*(A1)/150 ^d	Proportion of mineral fertilizers (really) replaced (%)			Amount of marginal mineral fertilizers avoided (kg/t material)		
	(A1)	(A2)	(A3)		(C1) ^e	(C2) = 23 ^d /(A2)*(B)	(C3) = 61 ^d /(A3)*(B)	(D1) = (A1) * (C1)	(D2) = (A2) * (C2)	(D3) = (A3) * (C3)
	N	P	K		N	P	K	N	P	K
Raw pig slurry	5.03	1.19	2.83	0.026	75%	50%	55%	3.8	0.59	1.56
Liquid fraction	2.91	0.24	1.92	0.013	65%	>100% ^f	41%	1.9	0.24	0.78
GW compost	5.82	1.34	12.21	0.013	40%	27%	22%	2.3	0.31	0.82
Digestate, maize scenario	5.16	1.11	3.77	0.026	75%	55%	42%	3.9	0.61	1.60
Digestate, straw scenario	5.38	1.23	4.72	0.027	75%	52%	35%	4.0	0.63	1.66
Digestate, HW scenario	7.12	1.34	3.03	0.036	75%	63%	73%	5.3	0.84	2.20
Digestate, CW scenario	4.96	1.15	3.78	0.025	75%	51%	41%	3.7	0.58	1.53
Digestate, GW scenario	7.33	1.24	2.61	0.037	75%	70%	87%	5.5	0.86	2.27
Digestate, SS scenario	9.73	3.67	3.86	0.049	75%	31%	78%	7.3	1.15	3.01
Digestate, mono-digestion	5.18	1.23	2.88	0.026	75%	50%	56%	3.9	0.61	1.60
Digestate, maize10% scenario ^b	8.46	1.34	5.31	0.043	75%	55%	42%	6.3	0.73	2.25
Digestate, straw scenario (alkali) ^b	5.03	1.14	4.68	0.026	75%	52%	33%	3.8	0.59	1.56
Digestate, straw scenario (enzymatic) ^b	5.16	1.17	4.99	0.026	75%	52%	32%	3.9	0.61	1.60

^a GW: garden waste; HW: Housesold biowaste; CW: Commercial biowaste; SS: Manure source-segregation

^b Sensitivity analysis scenarios.

^c These are as found in the inventory for each scenarios, section 3.

^d 150 kg N/ha; 23 kg P/ha and 61 kg K/ha needed by the crop rotation, as presented in Table S63.

^e Defined by law [67]. For the liquid fraction, the value for "urine" was considered. For compost, the value for "other types of organic fertilizers" was considered. For all digestates, the law mentions that the same efficiency as raw pig manure can be considered, so this was applied for the digestates in this study.

^f This means that fertilizer is missing to fully fulfill the crop requirements in P. Therefore, there is no excess, and 100% of the P applied does correspond to mineral P avoided.

In Table S65, the amount of N and P applied per FU for each digestate is presented.

Table S65. Nutrients applied on land per FU due to application of the organic materials on land

“Material” applied on land	N applied (kg/FU)	P applied (kg/FU)	K applied (kg/FU)
Raw pig slurry ex-storage (1 tonne)	5.1	1.2	2.9
Liquid fraction	22.9	1.9	15.1
GW compost	0.7	0.2	1.5
Digestate, maize scenario	10.6	2.3	7.7
Digestate, straw scenario	5.9	1.4	5.2
Digestate, household biowaste scenario	11.9	2.2	5.0
Digestate, commercial biowaste scenario	18.5	3.1	6.6
Digestate, garden waste scenario	6.1	1.4	4.7
Digestate, manure source-segregation scenario	22.0	8.3	8.7
Digestate, mono-digestion	5.1	1.2	2.9
Raw manure from farm 2 (if not used for biogas), manure source-segregation scenario	38.9	9.2	21.7

7. Modelling the nutrient losses (N and P) during application on land

Losses of P to soil and water were considered to correspond to 5% of the P applied in excess, based on [1]. 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 amount of P applied in excess (or surplus), can be calculated as the difference between the P applied, and the P uptake by the crop rotation. To transform the P uptake from (kg/ha) to (kg/ton applied), the area needed to apply the digestate, as calculated in column B of Table S66, can be used. The P uptake from the crop rotation is presented in Table S67.

Table S67. P uptake from the crop rotation

Year	Crop	P uptake (kg ha ⁻¹) ^a
1	Winter barley	14.54
2	Winter rape	19.11
3	Winter wheat	15.71
4	Winter wheat	15.71
5	Spring barley & catch crop	12.75
6	Spring barley	12.75
Annual average		15.09

^a Data from [9]. The P uptake from catch crops is not accounted for, as the catch crop biomass is completely returned to soil.

Leaching of N was calculated with the N-LES₄ model [15], a continuously updated empirical model to predict N leaching from arable land based on 1200 leaching studies performed in Denmark during the last 15 years. The resulting N and P losses during the application on land of the digestates (and other organic materials) is presented in Table S68.

Table S68. N and P losses due to application of the organic materials on land

“Material” applied on land	N leach from N-LES ₄ (kg/ha)	N leach used in this study (kg/t material)	P losses (kg/t)
Raw pig slurry	65.8	1.684	0.0402
Liquid fraction	71.3	0.914	0.0025
GW compost	109.2	1.469	0.0473
Digestate, maize scenario	65.8	1.726	0.0357
Digestate, straw scenario	65.8	1.800	0.0409
Digestate, household biowaste scenario	65.8	2.382	0.0397
Digestate, commercial biowaste scenario	65.8	1.659	0.0339
Digestate, garden waste scenario	65.8	2.452	0.0385
Digestate, manure source-segregation scenario	65.8	3.255	0.146
Digestate, mono-digestion	65.8	1.733	0.0416
Digestate, maize10% scenario ^a	65.8	2.831	0.0345
Digestate, straw scenario (alkali) ^a	65.8	1.683	0.0377
Digestate, straw scenario (enzymatic) ^a	65.8	1.726	0.0387

^a Sensitivity analysis scenarios.

8. Digestates' carbon fate

In this study, changes in soil C were estimated with the dynamic soil C model C-TOOL, developed to calculate the soil carbon dynamics in relation to the Danish commitments to UNFCCC. This model is parametrized and validated against long-term field experiments conducted in Denmark, UK and Sweden. Further description of the C-TOOL model is given in [41,68]. As opposed to many different soil C model, C-TOOL does not only consider the topsoil, but the whole 0-100 cm profile.

The output from C tool gives the level of soil C, after the continuous application of the digestate, years after years, from year 0 to year 200. The initial soil C level (year 0) is considered at 143.92 t ha^{-1} , based on [16], for a sandy soil. An example is provided in Figure S8, with the case of the application of the digestate from the commercial biowaste scenario.

As it can be seen from Figure S8, the change in soil C is much more pronounced in the first years, and eventually levels out to reach a new equilibrium. When performing LCAs, a time perspective is needed to consider such changes. Ideally, the specific yearly releases should be considered year per year, over the temporal scope considered for the study (here 2020). In this study, the model is performed over a horizon time of 1 y (which can be repeated e.g. 8 years, if this is the temporal scope selected). For simplicity, the soil C changes were annualized over 20 years, in conformity with the IPCC [69], and with the Renewable Energy Directive [70].

The amount of CO_2 released to the atmosphere from the different digestates was thus calculated as the total amount of C applied, minus the amount entering the soil. This, as well as C-TOOL results, is presented in Table S69. It should also be noted that the curve presented in Figure S8 includes the input of above- and below- ground residues from the crop rotation, which was estimated to 2.731 t C/ha (applies for all scenarios), based on the values found in [16]. Without the residues, most scenarios would likely result in a decrease in soil C.

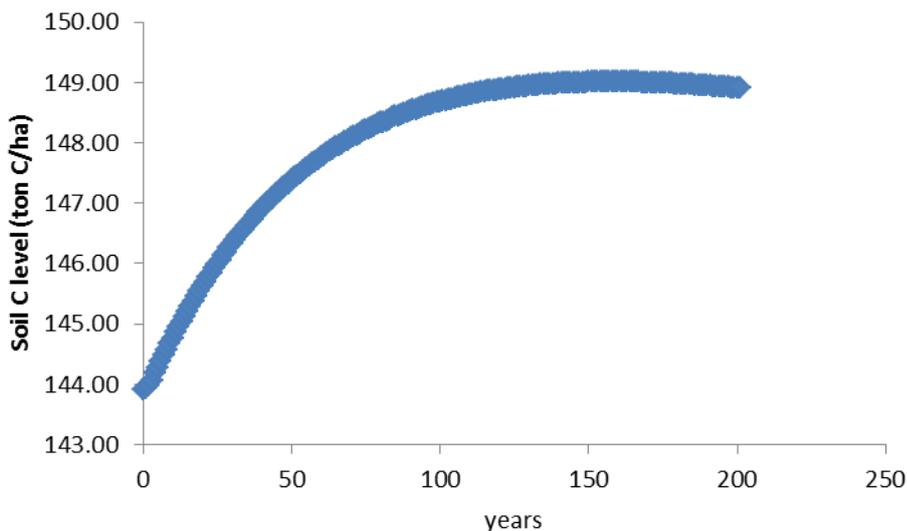


Figure S8. Example of output results from C-TOOL: case of the digestate from the commercial biowaste scenario

Table S69. C balance for the organic fertilizers applied on land

Parameter	Unit	Raw manure	Liquid fraction	GW compost	Digestate maize	Digestate straw	Digestate HW ^a	Digestate CW ^a	Digestate GW ^a	Digestate SS ^a	Digestate mono-digestion	Digestate straw alk. ^a	Digestate straw enz. ^a	Digestate maize-10% ^a
C in organic fertilizer	kg C/t applied	31.6	6.8	143.4	37.2	49.5	43.2	55.8	31.9	51.4	19.1	49.7	38.8	57.5
C to soil C pool ^b	kg C/t applied	0.53	-2.0	35.2	11.0	14.5	5.6	3.1	0.70	5.8	-2.7	5.2	6.0	2.0
C lost as CO ₂ -C ^c	kg CO ₂ -C/t applied	31.1	8.8	108.2	26.2	34.9	37.6	52.6	31.2	45.6	21.9	44.6	32.8	55.5
CO ₂ -C, as a % of C applied	%	98%	130% ^d	75%	71%	71%	87%	94%	98%	89%	114%	90%	84%	97%

^a HW: household biowaste; CW: commercial biowaste; GW: garden waste; SS: source-segregated manure; alk.: alkali pre-treatment; enz.: explosion-enzymatic pre-treatment; maize-10%: allowing for a mixture input reaching 10% DM in reactor.

^b Annualized over 20 years. Negative numbers represent a loss of soil C. This loss is assumed to entirely convert as CO₂-C to the atmosphere.

^c Corresponds to the C applied, minus C entering the soil C pool. It may be slightly underestimated, as the C entering the soil C is not only C from the digestate, but also from the 2.731 t C/ha above- and below-ground residues from the crop rotation.

^d This means that besides the C from the liquid fraction, some of the native soil C is also converted to CO₂.

9. Sensitivity analysis details

All sensitivity analyses have been performed for the global warming impact only. Therefore, the description below only focuses on the modelling of the GHG (and not the other substances) related to these sensitivity analyses.

9.1 Straw plowing instead of combustion

In this sensitivity analysis, the lost alternative for straw is considered to be straw plowing. This means that if the 0.190 t straw (per FU) (Figure S2) would not have been used for anaerobic co-digestion, it would have been incorporated in the soil instead. Part of the C of the straw would have entered the soil C pool, building up soil C stock, while most of it would have end up as a CO₂ emission to the atmosphere. Based on C-TOOL (see section 8), it was calculated that out of the 72.8 kg C applied with straw per FU (0.190 t straw /FU* 382.5 kg C/t straw, Table S24) 0.0115 kg C ha⁻¹ y⁻¹ (20 years annualization) is entering the soil C pool, while the remaining (i.e. 99.98% of the initial C in straw) ends up as CO₂-C. This CO₂-C emission is then avoided by avoiding straw incorporation (at the expense of anaerobic co-digestion).

9.2 Landfilling of the biowastes instead of combustion or composting

In this sensitivity analysis, it is considered that the lost alternative for household biowaste, commercial biowaste, and garden waste is landfilling instead of combustion (household and commercial biowaste) and composting (garden waste). A landfilling without recovery of the landfill biogas is assumed. The calculation of CH₄ emissions due to the landfilling of these biowastes was based on the IPCC guidelines [71], using a value of 1 for MCF, and 0.5 for DOCf (fraction of degradable organic C that can decompose), 0.5 for F (fraction of CH₄ in emitted landfill gas) and 0.1 for OX (oxidation factor). The values for these parameters were determined based on the values used in the latest Danish GHG report to the UNFCCC [3]. The 0.5 value for the DOCf parameter may be debated; in fact, [72] mention that it is rather 75% for food waste, and 64% for “average putrescible”, while [73] mention it is 100%. The calculated CH₄ emissions may therefore be seen as slightly underestimated. To report the calculated value in terms of annual release, an annualization over 20 years has been considered, in consistency with the annualization period used for soil C changes.

The CO₂ was estimated with the rough assumption that all the degradable C not converted to CH₄ is converted to CO₂. As for CH₄, an annualization over 20 years was assumed. The N₂O was assumed to be negligible, based on [72]. The calculated emissions of GHG during landfill are presented in Table S70 for all biowastes.

Table S70. GHG emissions due to the landfill of the biowaste. All values in g per tonne of biowaste per y.

	HW	CW	GW	Comment
Mass of degradable C deposited	70.9	63.6	81.6	Based on Table S58, and on the IPCC guidelines (DOCf = 0.5; MCF=1).
CH ₄ emission	2.13	1.91	2.45	Based on the IPCC guidelines (F = 0.5; OX=0.1). Annualized over 20 y.
CO ₂ emission	7.15	6.41	8.23	Assumed as 100% of the degradable C not emitted as CH ₄ . Annualized over 20 y.
N ₂ O emission	0	0	0	Assumed negligible, based on [72].

9.3 Mono-digestion instead of separation

This sensitivity analysis considers that the lost alternative for the manure source-segregation scenario is biogas (mono-digestion) instead of conventional manure management. The data used for modeling the mono-digestion are as in section 3.7. The results (per FU) are presented in Figure S9, where the changed flows appear in red.

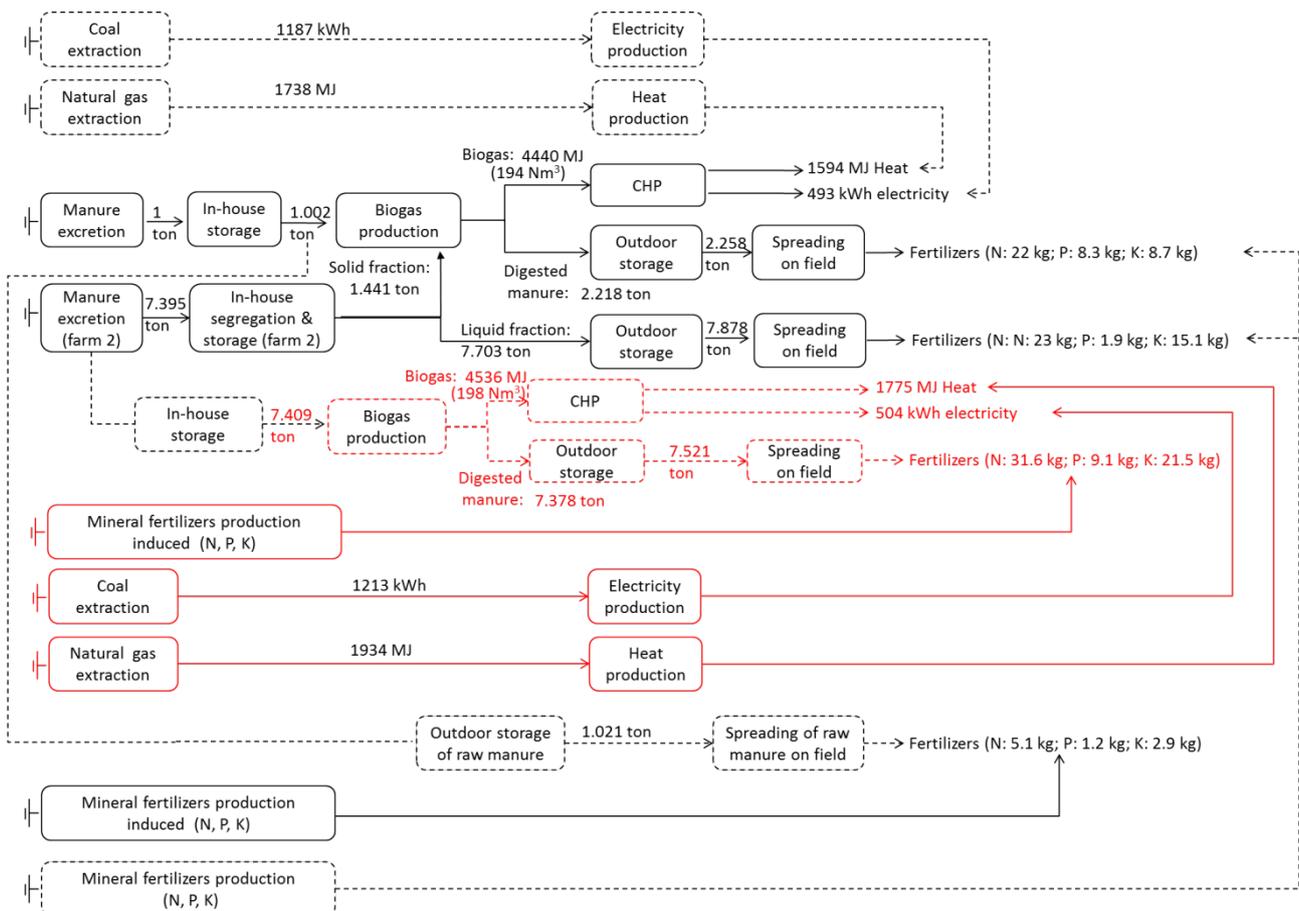


Figure S9. Process flow diagram for the sensitivity analysis considering anaerobic mono-digestion as the lost alternative in the manure source-segregation scenario. Dotted lines indicate avoided flows. Red flows indicate changed flows as compared to the baseline scenario.

9.4 Displaced crop for the maize scenario

For the maize scenario, the baseline scenario considers maize (for feed) as the displaced crop (or marginal crop) in Denmark for cultivating the maize silage needed for biogas. The difference, in terms of environmental impacts, between cultivating the new crop (maize silage) instead of the displaced crop (maize for feed, in the baseline scenario) represents the so-called direct land use change (DLUC). In the baseline case, the DLUC impact was assumed negligible. Yet, 2 sensitivity analyses were made with a different displaced crop, namely spring barley (with straw incorporation) and sugar beet (with top incorporation).

There are two main important points when considering a different crop displaced. The first is the difference in cultivation between the displaced crop and the maize-for-biogas, as above-described. The second relates to the ILUC.

For the first point, the inventory data considered for the cultivation of these displaced crops were taken from [16] (sandy soil, wet climate, "medium initial" soil C content). Key data for these crops are presented in Table S71.

Table S71. Key life cycle inventory data used for the cultivation of spring barley, maize silage and sugar beet.

	Spring barley	Maize silage	Sugar beet
Yield (t dm/ha)	4.25	12.05	12.45
CO ₂ -C in plant uptake (kg CO ₂ -C/ha*y)	-5087	-7497	-9094
CO ₂ -C biogenic (from above- and below-ground residues and manure input) (kg CO ₂ -C/ha*y) (20 y annualization)	3012	3083	3495
C in the biomass harvested (kg C/ha*y)	1913	5423	5603
N ₂ O-N in field (direct & indirect)	2.69	3.06	3.78
Net GWP for the whole cultivation process (kg CO ₂ eq/ha*y)	-1910	-13300	-15400

The CO₂-C from plant uptake consists of the C found in the harvested biomass, as well as the C lost in above- and below- ground residues. In the case of maize, the C harvested in the biomass is ultimately considered to return back to atmosphere, as the biogas is burned, and as the digestate is used on land. Therefore, in the case of the avoided spring barley/sugar beet, the return of the sequestered C that would have been harvested had to be considered as well. This effect is integrated in the DLUC bar of Fig 3b (from the main manuscript).

Regarding the ILUC point, the rationale is that when a crop is displaced, it would have provided a certain amount of DM to the food/feed market that is no longer available. When feed maize was displaced, 0.407 t DM were no longer available on the carbohydrate market (section 3.1.12). Based on the above yields, and on the 0.0338 ha of marginal crop displaced per FU (section 3.1.12), barley would have provided 0.144 t DM, and sugar beet 0.421 (although sugar beet in fact rather interacts on the sugar market). This indicates that the lower the yield of the displaced crop, the lower would have been the amount of crop DM provided on the market, and therefore the ILUC reaction to compensate for this is also expected to be lower. In section 3.1.12, it is described that an ILUC of 0.195 ha converted per ton DM displaced was considered, on the basis of Kløverpris' results [36]. This corresponded to 2.35 ha transformed/ha displaced, when applied

to feed maize as the crop displaced. When applied to barley and sugar beet respectively, this corresponds to 0.83 ha transformed/ha displaced and 2.43 ha transformed/ha displaced.

9.5 Pre-treatments for straw

The baseline scenario considers extrusion as the pre-treatment for straw. A sensitivity analysis has been made considering 2 others pre-treatments.

One is an enzymatic pre-treatment coupled with straw explosion. The first step consists of straw steaming at 200 ° C for 10 minutes with water to straw ratio 2:1 [74]. The second step consists of an enzymatic treatment of the straw “exploded”, assuming an enzyme load (Cellic CTec3 enzyme) of 24.0 kg enzyme per 1 000.0 kg straw and an energy consumption of 5.3 kWh/1 000.0 kg straw [75]. The degradability of straw was assumed to be doubled as a result of this pre-treatment (so a VS degradability of straw of 90% was assumed). The resulting changes in the flows are presented in Figure S10.

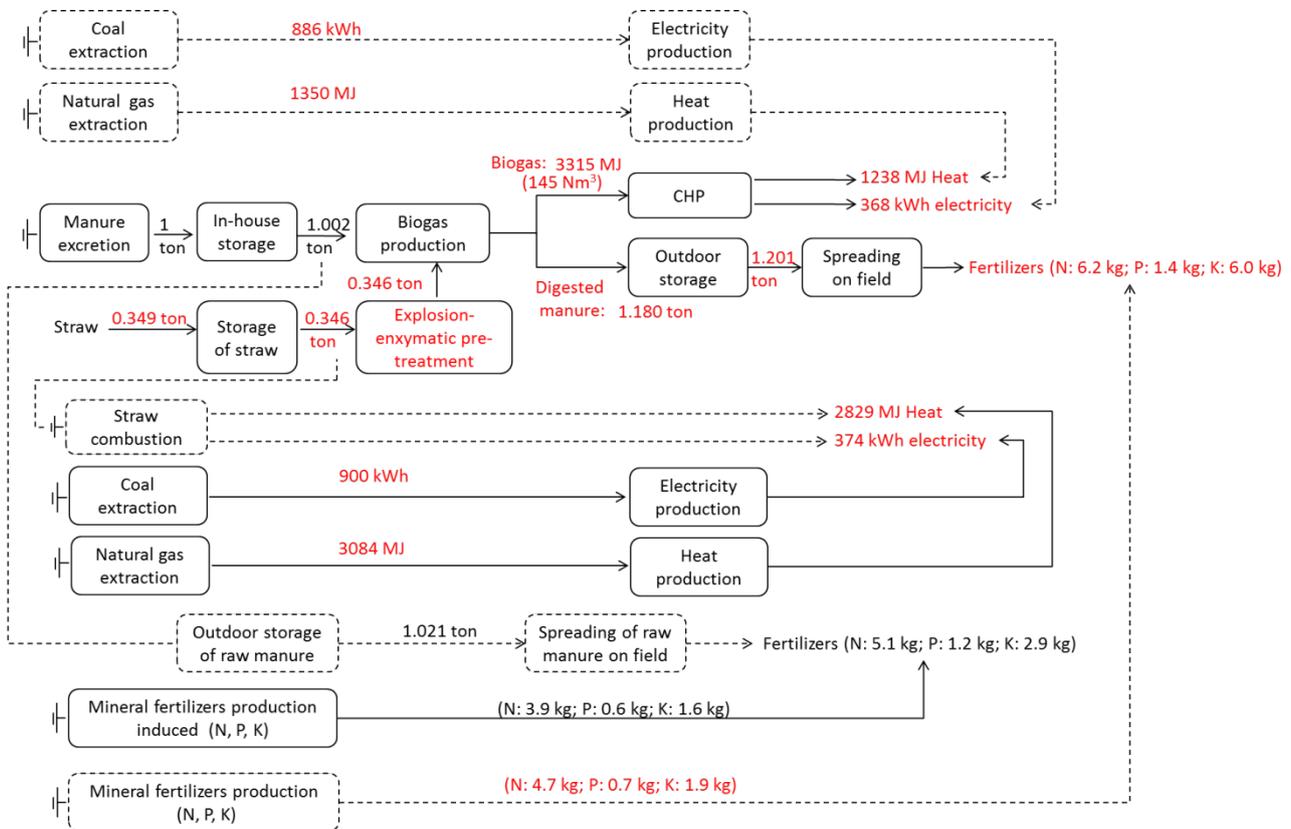


Figure S10. Process flow diagram for the sensitivity analysis considering a straw explosion combined with an enzymatic pre-treatment for the straw scenario (straw explosion combined with enzymatic hydrolysis). Dotted lines indicate avoided flows. Red flows indicate changed flows as compared to the baseline scenario.

The other pre-treatment considered is an alkali pre-treatment, performed at ambient temperature for 24 hours with 12.7 kg calcium hydroxide per kg of straw, at the concentration of 1.5 % DM, based on [76]. An electricity consumption of 7.5 kWh per t of straw was considered for cutting the straw [46] and of 5.3 kWh

for the treatment operation. A water addition of 384 kg per t of straw was also considered. Based on the laboratory tests performed in on [76], an increase in the CH₄ yield of 63% could be achieved. Assuming that lower yield would be obtained in “real-field conditions”, an increase of 40% only was considered in this study. The resulting changes in the flows are presented in Figure S11.

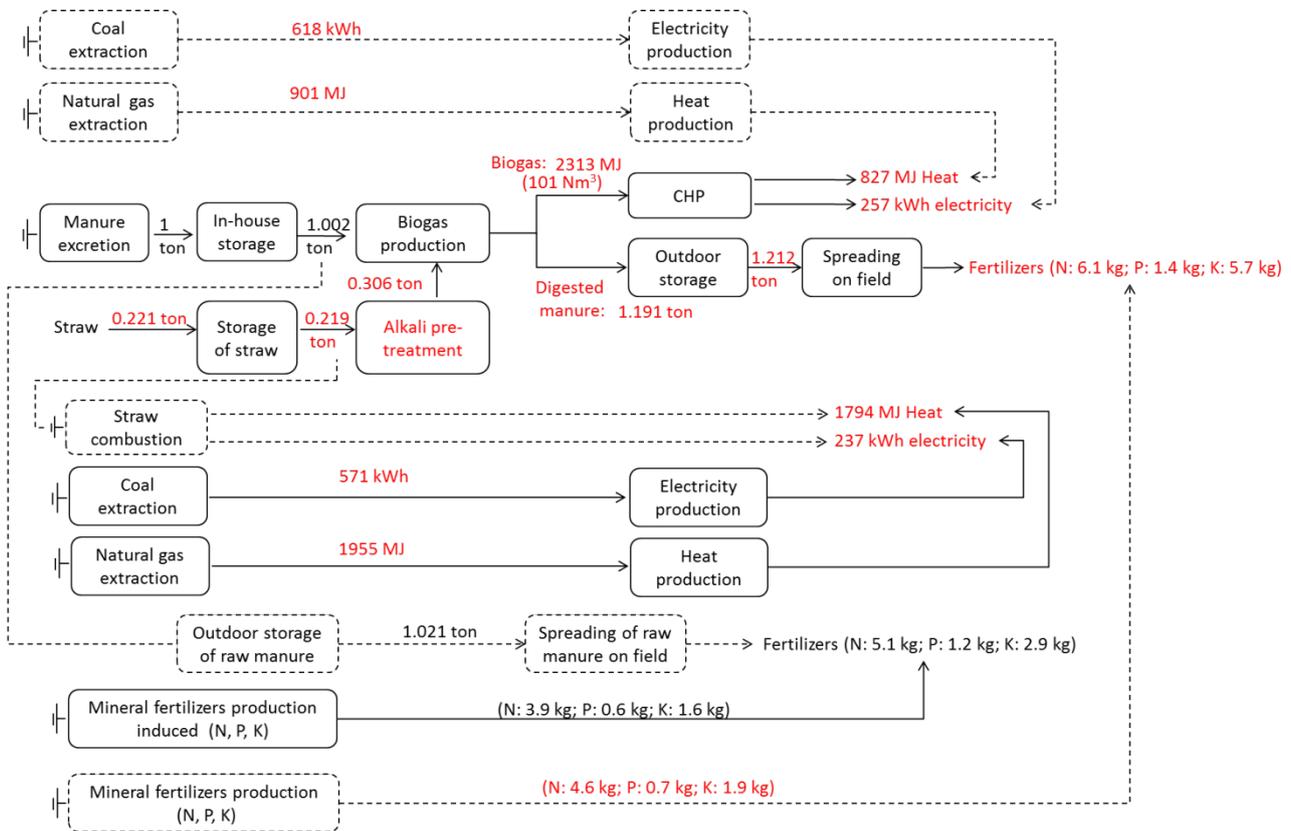


Figure S11. Process flow diagram for the sensitivity analysis considering an alkali pre-treatment for the straw scenario. Dotted lines indicate avoided flows. Red flows indicate changed flows as compared to the baseline scenario.

9.6 Natural drying of maize to reach 10% DM

In the baseline scenario for maize, it was not possible to reach the 10% DM in the input mixture that was used for the other scenarios (only 6.6% was reached, see Table 1 of the main manuscript). This was due to the low DM content of maize silage (31%). A natural drying of the maize on-field was thus considered in this sensitivity analysis, where the maize could reach a DM content of 40% (the emissions due to on-field drying were neglected). Based on this, the input mixture consisted of 25.8% manure ex-housing, and 74.2% maize silage (the maize share in the baseline was 57%, see Table 1 of the main manuscript). To limit the C/N ratio to 20, the addition of 1.6 kg N per t input mixture (here taken as calcium ammonium nitrate) was considered.

The resulting process flow diagram for this scenario is presented in Figure S12.

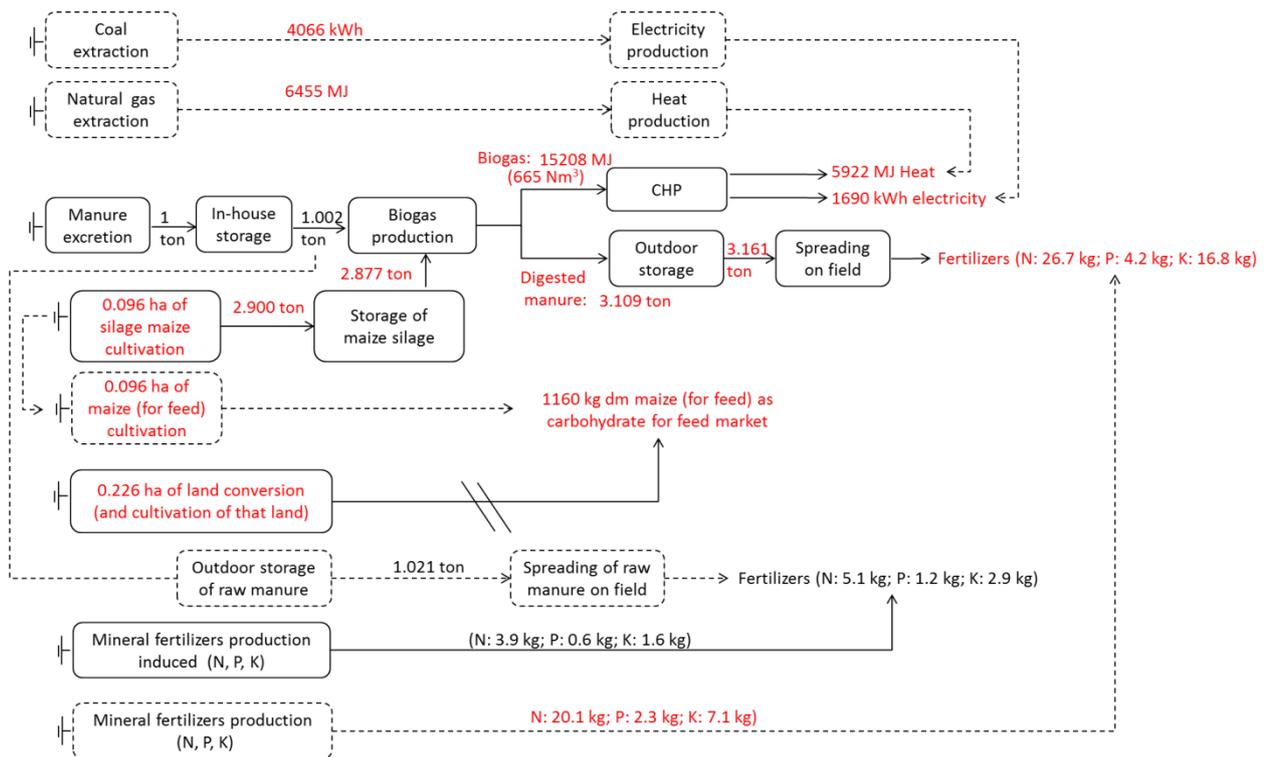


Figure S12. Process flow diagram for the sensitivity analysis considering a natural on-field drying of maize, so the biomass mixture in the digester can reach 10% DM (maize scenario). Dotted lines indicate avoided flows. Red flows indicate changed flows as compared to the baseline scenario. It should be noted that not all the converted land will be cultivated in feed maize, and that not all the maize (for feed) displaced is necessarily replaced, due to various market interactions.

10. Results' details (for global warming in the baseline and sensitivity analyses)

The detailed results (in terms of absolute values) are presented in Table S72-Table S76 for the global warming impact (expressed per FU, per Nm³ biogas produced and per tonne DM input). Table S72-Table S74 thus present the global warming results for the baseline case, while Table S75-Table S76 present the sensitivity analysis results on the lost alternatives and on the selected straw and maize scenario variations, respectively.

For each scenario, the “co-substrate lost alternative” consists of an aggregation of different processes. Figure S13 presents the breakdown of the different processes constituting the lost alternative, also for the case of the global warming impact.

Table S72. LCA results for global warming, in kg CO₂ eq. per FU (baseline scenario)^a

	Maize	Straw	HW	CW	GW	SS	Mono
Co-substrate lost alternative	1565	45	82	-38	-157	-1681	-
Reference manure handling avoided	-228	-228	-228	-228	-228	-228	-228
Avoided fertilizers	-61	-34	-69	-107	-35	-243	-30
Avoided heat (from biogas)	-174	-57	-86	-127	-47	-130	-12
Avoided electricity (from biogas)	-615	-213	-321	-474	-184	-478	-66
Biogas process	118	41	61	90	35	91	13
Digestate handling	349	237	375	737	244	624	138
Segregated liquid fraction handling	-	-	-	-	-	557	-
Others	65	67	85	115	59	232	58
Net	1018	-142	-101	-32	-313	-1256	-128

^a HW: Household biowaste; CW: Commercial biowaste; GW: Garden waste; SS: Source-segregated solid manure fraction; Mono: mono-digestion.

Table S73. LCA results for global warming, in kg CO₂ eq. per Nm³ biogas produced (baseline scenario)^a

	Maize	Straw	HW	CW	GW	SS	Mono
Co-substrate lost alternative	6.27	0.52	0.63	-0.20	-2.10	-8.66	-
Reference manure handling avoided	-0.91	-2.64	-1.75	-1.19	-3.06	-1.18	-8.51
Avoided fertilizers	-0.25	-0.40	-0.53	-0.56	-0.48	-1.25	-1.13
Avoided heat (from biogas)	-0.70	-0.66	-0.66	-0.66	-0.63	-0.67	-0.46
Avoided electricity (from biogas)	-2.46	-2.46	-2.46	-2.46	-2.46	-2.46	-2.46
Biogas process	0.47	0.47	0.47	0.47	0.47	0.47	0.47
Digestate handling	1.40	2.74	2.87	3.83	3.27	3.22	5.13
Segregated liquid fraction handling	-	-	-	-	-	2.97	-
Others	0.26	0.79	0.66	0.60	0.79	1.20	2.20
Net	4.08	-1.64	-0.77	-0.16	-4.19	-6.47	-4.76

^a HW: Household biowaste; CW: Commercial biowaste; GW: Garden waste; SS: Source-segregated solid manure fraction; Mono: mono-digestion.

Table S74. LCA results for global warming, in kg CO₂ eq. per t DM input (baseline scenario)^a

	Maize	Straw	HW	CW	GW	SS	Mono
Co-substrate lost alternative	3.34	0.20	0.26	-0.08	-0.63	-3.64	
Reference manure handling avoided	-0.49	-1.02	-0.72	-0.47	-0.91	-0.49	-3.32
Avoided fertilizers	-0.13	-0.15	-0.22	-0.22	-0.14	-0.52	-0.43
Avoided heat (from biogas)	-0.37	-0.25	-0.27	-0.26	-0.19	-0.28	-0.18
Avoided electricity (from biogas)	-1.31	-0.95	-1.02	-0.98	-0.74	-1.04	-0.96
Biogas process	0.25	0.18	0.19	0.19	0.14	0.20	0.18
Digestate handling	0.75	1.06	1.19	1.52	0.98	1.35	2.00
Segregated liquid fraction handling	-	-	-	-	-	1.21	-
Others	0.14	0.30	0.27	0.24	0.24	0.50	0.85
Net	2.17	-0.64	-0.32	-0.07	-1.26	-2.72	-1.85

^a HW: Household biowaste; CW: Commercial biowaste; GW: Garden waste; SS: Source-segregated solid manure fraction; Mono: mono-digestion.

Table S75. LCA results for the sensitivity analysis on the lost alternative, for global warming, in kg CO₂ eq. per FU^a

	Straw		HW		CW		GW			SS	
	bsl.	plo.	bsl.	ldf.	bsl.	ldf.	bsl.	inc.	ldf.	bsl.	m.d.
Co-substrate lost alternative	45	-268	82	-0.05	-39	-0.09	-157	18	-0.019	-1681	-496
Reference manure handling avoided	-228	-228	-228	-228	-228	-228	-228	-228	-228	-228	-228
Avoided fertilizers	-34	-34	-69	-69	-107	-107	-35	-35	-35	-242	-242
Avoided heat (from biogas)	-57	-57	-86	-86	-127	-127	-47	-47	-47	-130	-130
Avoided electricity (from biogas)	-213	-213	-321	-321	-474	-474	-184	-184	-184	-478	-478
Biogas process	41	41	61	61	90	90	35	35	35	91	91
Digestate handling	237	237	375	375	737	737	244	244	244	624	624
Segregated liquid fraction handling	-	-	-	-	-	-	-	-	-	557	557
Others	67	67	85	85	115	115	59	59	59	232	232
Net	-143	-456	-101	-184	-32	6.20	-313	-139	-157	-1256	-71

^a HW: Household biowaste; CW: Commercial biowaste; GW: Garden waste; SS: Source-segregated solid manure fraction; bsl: baseline; plo.: plowing; ldf.: landfilling; inc.: incineration; m.d: mono-digestion.

Table S76. LCA results for the sensitivity analysis on the variants for the straw and maize scenarios, for global warming, in kg CO₂ eq. per FU ^a

	Maize				Straw		
	bsl.	10%+N	barley DLUC	s.beet DLUC	bsl.	alk.	enz.
Co-substrate lost alternative	1565	4285	552	1618	45	52	82
Reference manure handling avoided	-228	-228	-228	-228	-228	-228	-228
Avoided fertilizers	-61	-154	-61	-61	-34	-35	-36
Avoided heat (from biogas)	-174	-482	-174	-174	-57	-67	-101
Avoided electricity (from biogas)	-615	-1637	-615	-615	-213	-249	-357
Biogas process	118	359	118	118	41	47	68
Digestate handling	349	1017	349	349	237	301	245
Straw handling & pre-treatment	-	-	-	-	9	11	188
DLUC	-	-	-140	767	-	-	-
Others	65	73	65	65	58	58	58
Net	1018	3233	-135	1838	-143	-109	-80

^a bsl: baseline; s.beet: sugar beet; alk: alkali pre-treatment; enz: explosion-enzymatic pre-treatment

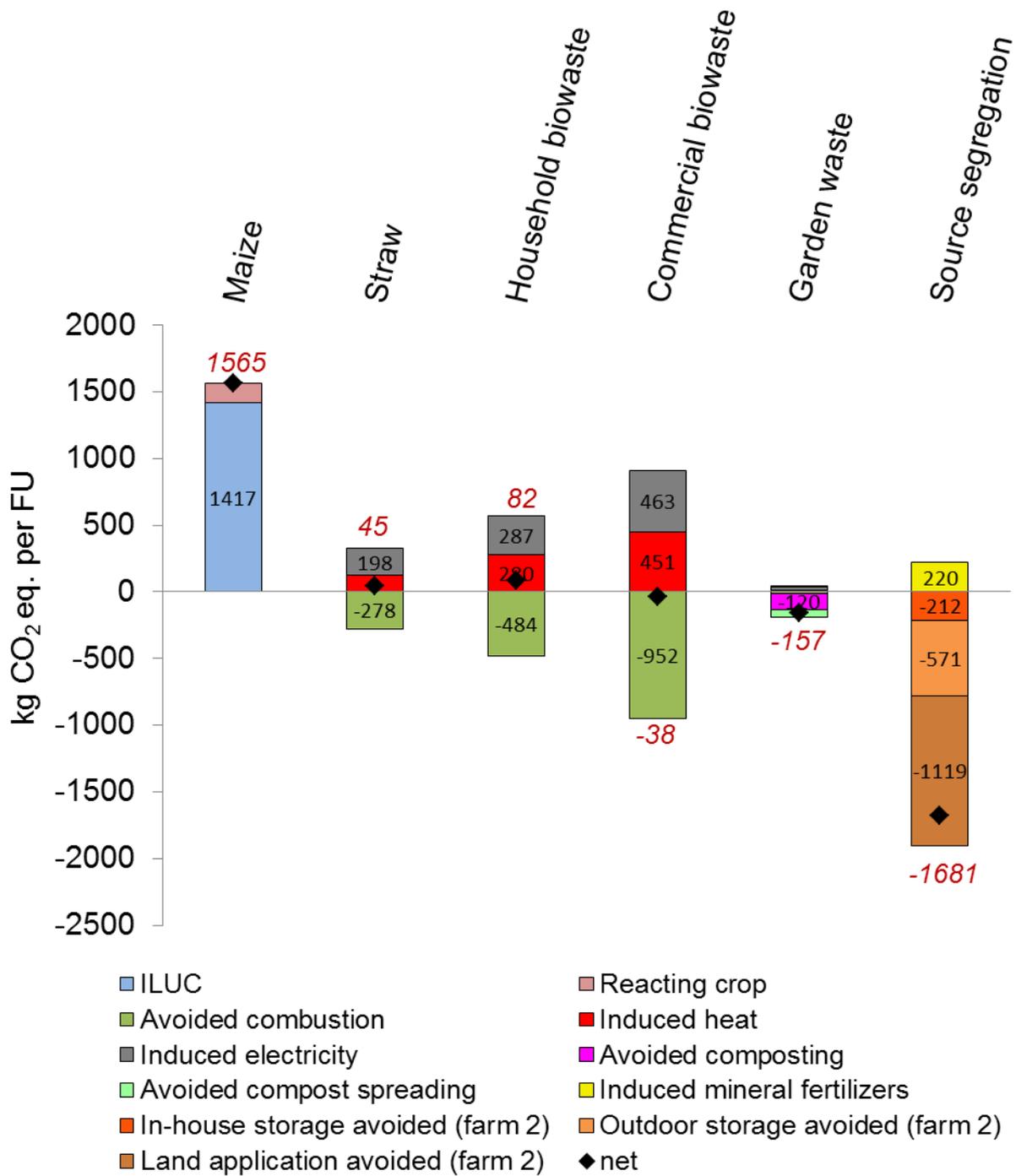


Figure S13. Breakdown of the “co-substrate lost alternative” impact, for the baseline scenarios (global warming impact). Net values are indicated in red, at the top (if positive) or bottom (if negative) of the bars.

11. Calculation of ILUC figures per energy unit

Two main types of ILUC figures are mentioned in this study; i) the area expanded per ha of crop displaced; and ii) the amount of CO₂ eq (here expressed in tonnes) per ha of crop displaced.

In both cases, our results are expressed per ha of crop displaced, which is the most appropriate in the case of this study. However, as most ILUC studies have been carried out in the context of biofuel mandates, figures are typically reported per MJ of energy in the fuel (before combustion losses). In order to compare with these figures, our results were converted per unit of energy. However, it should be noted that our results do not directly compare with those derived from these biofuels study, as their ILUC figures are an aggregation of multiple parameters (for example, the by-product of biofuels is assumed to replace feed, and the ha no longer needed to produce this feed are deducted from the ILUC figure derived).

In case i), the figure found in this study for the baseline case was 2.35 ha expanded per ha displaced (section 3.1.12). As 0.034 ha are converted per FU (Figure S1), and as the energy in the biogas produced (before conversion losses) is 5716 MJ (Figure S1), the conversion is as follows:

$$\frac{2.33 \text{ ha expanded}}{\text{ha displaced}} \times \frac{0.034 \text{ ha displaced}}{\text{FU}} \times \frac{\text{FU}}{5716 \text{ MJ}} \times \frac{1000 \text{ MJ}}{\text{GJ}} = \frac{0.0139 \text{ ha expanded}}{\text{GJ}}$$

In case ii), a figure of 357 ton CO₂ eq. per ha displaced was found, which was annualized over 20 y. The resulting conversion is thus:

$$\frac{357 \text{ t CO}_{2\text{eq.}}}{\text{ha displaced}} \times \frac{1}{20 \text{ y}} \times \frac{0.034 \text{ ha displaced}}{\text{FU}} \times \frac{\text{FU}}{5716 \text{ MJ}} \times \frac{1000 \text{ kg CO}_2}{\text{t CO}_2} \times \frac{1000 \text{ g CO}_2}{\text{kg CO}_2} = \frac{106 \text{ g CO}_{2\text{eq.}}}{\text{MJ y}}$$

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