Environmental assessment of renewable energy scenarios towards 2050

Coherent Energy and Environmental System Analysis Background Report Part 5

Astrup, Thomas; Tonini, Davide; Hamelin, Lorie; Wenzel, Henrik

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Coherent Energy and Environmental System Analysis

Background Report Part 5

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Authors:
Thomas Astrup, DTU Environment
Davide Tonini, DTU Environment
Lorie Hamelin, University of Southern Denmark
Henrik Wenzel, University of Southern Denmark

Publisher:
Department of Development and Planning
Aalborg University
Fibigerstræde 13
9220 Aalborg Ø
Denmark

Cover photo: Kristen Skelton

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Preface

This report presents a summary of results pertaining to environmental assessment of future energy systems of the strategic research project “Coherent Energy and Environmental System Analysis” which was conducted in 2007-2011 and funded by the Danish Council for Strategic Research together with the participating parties.

The project was interdisciplinary and involved more than 20 researchers from seven different universities or research institutions in Denmark. Moreover, the project was supported by an international advisory panel.

The work was carried out as an interaction between five work packages. In this work package on environmental assessment of future energy systems, researchers from the Department of Environmental Engineering (Technical University of Denmark) and the Institute of Chemical Engineering, Biotechnology and Environmental Technology (University of Southern Denmark) participated.

A number of reports, papers and tools were reported separately from each part of the project. A list of the background reports is given at the end of this preface while a complete list of all papers and reports can be found at www.ceesa.dk

This report details the articles and papers that were written specifically for this work package.

List of background reports:

Background Report Part 1: CEESA 100% Renewable Energy Scenarios towards 2050
Background Report Part 2: CEESA 100% Renewable Energy Transport Scenarios towards 2050
Background Report Part 3: Electric power systems for a transition to 100% renewable energy systems in Denmark before 2050
Background Report Part 4: Policies for a Transition to 100% Renewable Energy Systems in Denmark Before 2050
Background Report Part 5: Environmental Assessment of Renewable Energy Scenarios towards 2050

November 2011

Thomas Astrup
Work package coordinator, Work Package 5 – Environmental Assessment of Renewable Energy Scenarios towards 2050

Summary

The objective of Work Package 5 (WP5) is to carry out environmental assessment, namely consequential life cycle assessment (LCA), on the renewable energy scenarios defined by the other work packages.

Given the importance of the environmental consequences of biomass use, as compared to other renewable energy sources, the work carried out in this work package put a particular emphasis on the different implications of biomass use for energy. This includes land use changes implications, assessment of the biomass potential in Denmark, and the assessment of different conversion routes of biomass to energy.

The work has been done in collaboration with WP1 with respect to the definition of the energy system and WP2 with respect to biomass conversion technologies and identification of marginal products with importance to the system.

1. Introduction

The aim of this project is the integration of energy and environmental system analysis. Therefore, the outputs of the energy system analyses (WP1) are here used as inputs for the LCA study. The life-cycle assessment provides a comprehensive overview of the main potential environmental impacts associated with the future addressed energy systems and provides inputs for discussion and further improvement of the same energy systems.

The objectives of WP5 can be summarized as follows:

- Assessment of biomass potential for Denmark. This was done in collaboration with WP1 and WP2.
- Assessment of land use changes implications.
- Identification of marginal processes and activities with importance to the energy system. This was done in collaboration with WP2.
- Review and choice of biomass conversion technologies. This was done in collaboration with WP1 and WP2.
- Identification of primary and secondary services provided by the energy system. This was done in collaboration with WP1.
- Assessment of environmental impacts associated with the energy systems.

2. Biomass potential

The biomass resource potential for Denmark was assessed through an extensive literature review (Article 2). The collected data are shown in Table 1. The most abundant biomass resources for Denmark consisted of lignocellulosic materials (such as straw and wood) and manure. The total potential (excluding energy crops) was estimated in the range of about 176-184 PJ.

Since the total amount of biomass potential resources did not match the heat and electricity demand for the future scenarios (as well as the fuel demand for transport), cultivation of energy crops had to be included in the assessment. About 60 PJ of willow were estimated to be required in order to satisfy electricity and heat demand, based on the outputs of Mathiesen et al. 2010. According to Joergensen et al. (2008), the land «lying fallow» available for cultivation of lignocellulosic energy crops (e.g. willow) equaled about 9.1 PJ. Therefore, it was assumed that the remaining 51 PJ were cultivated at the expense of the marginal crop (spring barley, according to Weidema et al. 2003).

Biomass (PJ)	Foedevareministeriet (2008)		Energistyrelsen (2009)		Joergensen et al. (2008)	This study
	U	P	U	P	P	P
rapeseed	3.4	4.5	-	-	4.5	4.5
willow	0.5	9.1	-	-	9.1	9.1
grass[1]	0	5.1	-	-	5.1	13
straw	18.5	26.8	17.3	26.8	33.5	39
beet top	-	-	-	-	0.2	-
animal manure[1]	1.1	20.2	-	-	20.2	27
fiber fraction	0	2.5	-	-	2.5	2
mill residues	-	-	-	-	0.9	-
beet pulp	-	-	-	-	1.7	-
molasses	-	-	-	-	1.2	-
potato pulp	-	-	-	-	0.3	-
brewer's grain	-	-	-	-	0.6	-
whey	-	-	-	-	2.8	-
wood chips	-	-	9.8	40	7.7	9.8
fire wood	-	-	23		26	23
unexploited forest increment	-	-	-		17	-
wood pellets	-	-	2.3		2.6	2.3
wood residues	-	-	5.6		6.3	5.6
waste	-	-	23	34-41	-	47
paper and cardboard	-	-	5-6	5-6	-	-
industrial waste	0.9	1.5	-	-	-	-
animal fat	1.9	3.2	-	-	-	-
meat and bones	0	1.6	-	-	-	-
Total						182.3

Table 1 Biomass resource for Denmark. The values are expressed as primary energy (LHV) before energy conversion (except manure and grass whose potential is expressed as energy in the biogas)

3. Land use

3.1 Direct land use change (dLUC)

In a 100 % renewable energy system, biomass is the obvious storable fuel which thereby plays a key role in balancing the electricity system, at least for the transition from a fossil to a non fossil energy system. Moreover, as opposed to e.g. hydrogen, it can be converted into energy dense fuels (on a volumetric basis), and as such is likely to represent the key for aviation fuels (Wenzel, 2010). These properties make biomass an integral part of a 100 % renewable energy system.

Energy crops, as opposed to other biomass types such as animal manure or municipal waste, are not a co-product of another activity and as such are able to respond to an increase in demand of biomass-for-energy, meaning they are a key player of the biomass share of a fossil free energy system. However, because the amount of land that can be available for cultivation is fixed, using energy crops as part of the Danish energy strategy involves changes in the land use allocation in Denmark.

The term direct land use changes refers specifically to this change in the land use allocation, occurring as an immediate result of allocating more land to bioenergy production in a given country. Because the way land is used is a key parameter in the resulting biogeochemical flows of carbon (C) and nitrogen (N), any changes in the land use practices induce perturbations in the exchange of C and N between the land and the atmosphere. Because of the potential importance of energy crops into renewable energy systems, these changes must be thoroughly understood so the environmental consequences of different bioenergy crops systems can be highlighted to provide a rigorous scientific background for policy decisions to be made.

However, few data are available to fully consider the perturbations of these flows in LCA, in order to perform a complete and holistic substance balance, particularly for greenhouse gas (GHG), ammonia (NH_3) and phosphorus (P), in the case of bioenergy. This lack of LCA data applies particularly to the three following main points:

- Repartition of biomass above and below ground, and consequently the repartition of the C and N contained in it between these different pools. This is very seldom taken into account in LCA, and often completely ignored (Larson, 2006), although the need for it is more and more recognized and claimed (e.g. Cherubini et al., 2009; Brandao et al., 2010), especially in the light of including soil C changes into LCA. This is also very relevant for soundly including residue management into the picture and the changes induced by e.g. harvesting straw or other residue types from the different energy crops.

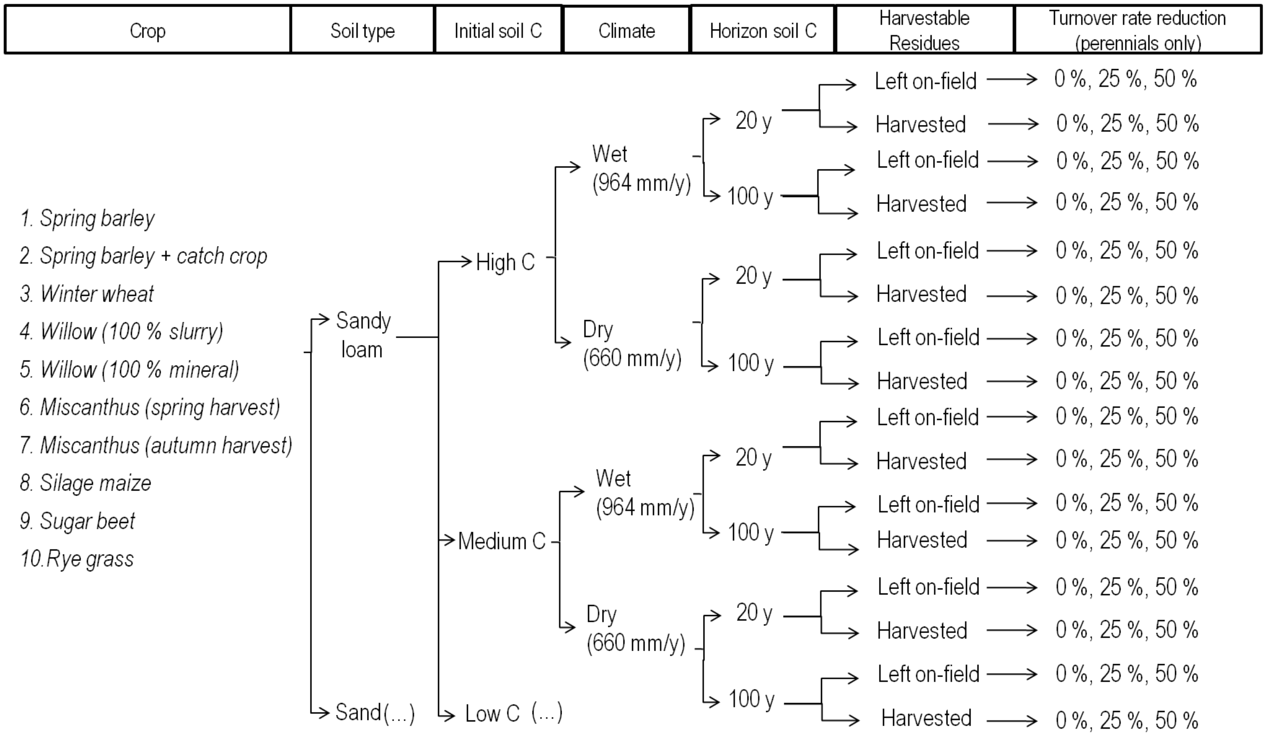
- Perennial crops. Although comprehensive LCA inventories do exist for some annual crops (e.g. Nemecek and Kägi, 2007; Jungbluth et al., 2007), very few complete LCA datasets, if any, are available for perennials like miscanthus and willow, albeit datasets do exist for some grass types. Given that both miscanthus and willow are envisioned to be key energy crops for a 100 % renewable energy system in Denmark, complete LCA datasets for these crops appear essential to fully judge the consequences of implementing these energy crops instead of others.

- Translating the numerous dynamic processes involved in bioenergy systems into a set of discrete values, as required for performing LCA. In a dynamic system such as the bioenergy one, many processes exhibiting a high degree of spatial and temporal variability are involved, and their translation to static values represents an important methodological challenge. Moreover, because of the importance of site-specific conditions (e.g. climate, soil type, initial soil C levels) on the biogeochemical flows of C and N involved in bioenergy systems, a comprehensive LCA database

> considering a high level of such site-specific details would be highly relevant for providing a strong background for decision-making regarding the bioenergy systems to prioritize in a 100 % renewable energy system.

In the light of these considerations, a comprehensive life cycle inventory database has been built in this project, based on complete state-of-the-art agro-ecological balances. This inventory also includes soil C balances which were performed with the 3-pooled dynamic soil model C-TOOL (Petersen et al., 2002; Petersen, 2007) used to calculate the soil C dynamics in relation to the Danish commitments to the United Nations Framework Convention for Climate Change (UNFCCC). The structure of this inventory is illustrated in Figure 1. A considerable level of detail has been included in the inventory, resulting in a total of 576 combinations. This includes 9 different crops, 2 soil types (clay and sand), 3 levels of initial C content in the soil (high, average, low), 2 climate types (wet and dry), 2 time horizons for soil C turnover (20 years and 100 years), 2 management possibilities for the residues (removal and incorporation into soil) as well as 3 soil carbon turnover rate reductions for perennial crops in response to the absence of tillage (0, 25 %, 50 %). The objective of this inventory is to provide accurate consequential data upstream the harvest of biomass, which can be used as a valuable input for assessing the further different fates of the harvested biomass. A consequential database means that an effort has been made to identify the marginal suppliers involved in the different processes, that is to say those responding to an incremental change in demand in the concerned markets. In the context of this study, this particularly applies to energy and fertilizers. Such an LCA consequential database is a clear innovative aspect of this study.

This life cycle inventory is the object of an article to be submitted to Global Change Biology Bioenergy by Hamelin et al., which is currently under redaction. As required by ISO standards on LCA (ISO 2006a; ISO 2006b), such inventory must be transparent and all assumptions and methodology choices to build it must be thoroughly explained. This has been done in the comprehensive inventory report enclosed with this report, see section 9.4.



Combinations detail:
Spring barley (1,2), winter wheat (3) & sugar beets (9): 4 crops x 2 soil x 3 initial soil C x 2 climate x 2 horizon time x 2 residues management = 192 combinations
Miscanthus (6,7) & willow (4,5): 4 crops x 2 soil x 3 initial soil C x 2 climate x 2 horizon time x 1 residues management x 3 turnover rates = 288 combinations
Rye grass (10): 1 crop x 2 soil x 3 initial soil C x 2 climate x 2 horizon time x 1 residues management x 3 turnover rates = 72 combinations
Silage maize (8): 1 crop x 2 soil x 3 initial soil C x 2 climate x 2 horizon time x 1 residues management = 24 combinations
Total = 576 combinations

Figure 1. Overview of the life cycle inventory database built in this project

3.2 Indirect land use change (iLUC)

Recent studies showed that an increased demand for bioenergy would lead to an overall increase in absolute global cropland requirements (e.g. Searchinger et al., 2008; Fargione et al., 2008; Kløverpris et al., 2008; Hertel et al., 2010; Tyner et al., 2010). This refers to the so-called indirect land use changes (iLUC), i.e. the land use change response which occurs when existing agricultural land previously used for food or feed production is devoted to the production of bioenergy feedstocks. In a nutshell, the rationale is that the resulting drop in the supply of feed or food causes a relative increase in agricultural prices, which then provides incentives to increase the production elsewhere (Kløverpris et al., 2008). Although a share of this production increase may be met by intensification (i.e. increased yield), the remaining share will come from conversion of new land to agricultural land, the result of which is potentially rather tremendous greenhouse gas (GHG) implications. This is especially true when lands rich in carbon content, such as tropical land, are converted to agricultural production. In fact, tropical ecosystems store about 340 billion tonnes of carbon, which is equivalent to more than 40 times the total annual anthropogenic emissions from fossil fuel combustion (Gibbs et al., 2008). Croezen et al. (2010) report an average value of 60 g CO_2 eq. MJ^{-1} of fuel produced (for a 20 year annualization) to reflect the GHG implications of iLUC only. This, however, should be seen as a low value, as higher values were reported by several other studies (e.g. Ros et al., 2010; Overmars et al., 2011; Edwards et al., 2010; Searchinger et al., 2008). Moreover, based on the results from Edwards et al. (2010), the exact GHG implications appear to be dependent on the direct land use change that happened in the first place (i.e. the energy crop type that is cultivated at the expense of food as well as where this happens).

There are many reasons to believe that the average value of 60 g CO_2 eq. MJ^{-1} of fuel produced proposed by Croezen et al. (2010) may be an underestimation. In fact, this value relies on the different published studies on iLUC, but most models published so far do not go fully downstream, i.e. the iLUC response may be that a new crop has to be established on grassland, but models do not include what was formerly done on that grassland (e.g. cattle grazing), and how this is compensated for (e.g. deforestation of new areas). This means that significant GHG releases are not included. For example, as demonstrated by Cederberg et al. (2011), the average GHG emissions of producing Brazilian beef in newly deforestated areas of Amazonia are 726 kg CO_2 eq. kg^{-1} carcass weight, which is 16.5 times higher than the GHG emissions of producing an average Brazilian beef (also including land use change emissions). Moreover, these models do not account for the N_2O emissions occurring due to intensification and the use of additional N fertilizer to boost the yield. As N_2O is a GHG with a considerable global warming potential (298 kg CO_2 equivalent per kg N_2O for a time horizon of 100 years; Forster et al., 2007), this intensification effect is likely to influence significantly the GHG overall response related to iLUC. Finally, most models do not include the emissions occurring as a result of tropical peat oxidation when cultures need to be established on undisturbed peat, which is also likely to be rather significant.

It is worth mentioning that even if the average iLUC emissions of 60 g CO_2 eq. MJ^{-1} of fuel produced proposed by Croezen et al. (2010) may be an underestimation, it is still enough, when added to the GHG from cultivation, processing, transport and distribution of the biofuels, to prevent any biofuels from energy crops to comply with the EU Renewable Energy Directive 2009/28/EC. This directive states that a GHG reduction of 35 % compared to fossil fuels should be obtained (which emit an average of 83.8 g CO_2 eq. MJ^{-1} of fuel).

For this project, the ambition is to identify the iLUC response of allocating more land to energy crops in Denmark and size the environmental consequences of this response. To relate the environmental impacts from iLUC to the crops displaced in Denmark, it is necessary to:

i. Identify the crop displaced in Denmark
ii. Estimate the amount of land affected by iLUC as well as the type of land affected (biome type, vegetation type before the conversion, e.g. forest or grassland)
iii. Estimate the resulting changes in C and N flows from land to atmosphere.

This work is currently under progress, and is to be the object of an article by Hamelin and Wenzel. The interest of this work lies in the quantification of the difference in environmental consequences (mostly GHG emissions) between the reference case (i.e. no crop displacement in Denmark) and the bioenergy case (i.e. Danish crops are displaced at the expense of energy crops, and compensated for somewhere else).

The main aspects to be tackled by this study are related to points i, ii and iii above:

i. **Identifying the crop displaced in Denmark**. This consists in identifying the existing uses of Danish land (e.g. pastures or agricultural cropland for food) that would be displaced at the expense of the energy crops. The crop to be displaced in Denmark is likely to be the less competitive one. Different LCA carried out in Europe identified, based on Weidema (2003), spring barley as the crop displaced by an increased demand for other crops (e.g. Schmidt, 2007; Dalgaard, 2007). This is because Weidema (2003) highlights spring barley as the crop with the lowest gross margin. However, this is based on statistics from 1992 and 1997. Recent outlook from Ireland (O'Mahony and O'Donovan, 2010; Clancy and Thorne, 2010) and from the UK (HGCA, 2010) tends to confirm spring barley as the crop with the lowest gross margin. Assuming spring barley as the displaced crop also makes sense from an agronomic point of view, since it is consistent with the assumption that displaced cropland (from food market to energy market) comes from the lower quality soils (i.e lower productivity land), leading to soils where spring barley is actually cultivated. This is why spring barley, as a potential displaced crop, has been included in the database described in section 3.1. Similarly, winter wheat and grassland were identified by St.Clair et al. (2008) as potential land uses to be displaced in the UK. These crops were also included in the database described in section 3.1. This study will therefore consider spring barley as the marginal (or displaced crop), but sensitivity analyses are to be carried out with winter wheat.

ii. **Estimate the amount of land affected by iLUC as well as the type of land affected.** This will be done through the use of a response model. Different approaches exist (risk based, chain analysis based on agricultural statistics and economical modeling, whether based on partial or general equilibrium) and these have been the object of recent publications (e.g. Croezen et al., 2010; Witzke et al., 2008; Edwards et al., 2010). The aim is to select a few of these approaches and compare the obtained results.

iii. **Estimate the resulting changes in C and N flows from land to atmosphere**. This involves the estimation of C in the vegetation of converted land as well as in the soil. Different databases and other data sources have been identified to this end (e.g. German WBGU carbon stock data, Woods Hole research centre dataset). This task also implies the quantification of the proportion of this C that will be released from these pools (i.e. vegetation and soil) to the atmosphere. It also involves the estimation of the opportunity cost for the sequestrating capacity lost, in cases where, for example, young forests are converted to agricultural production. The environmental consequences of the agricultural production on this new land will also be dealt with, as well as the time issue (i.e. the annualization of the GHG emissions over time).

3.3 Marginal fertilisers

If more crops are to be cultivated as a result of a 100 % Danish renewable energy system (in Denmark and beyond), more fertilizer will be used, and it will be the marginal fertiliser that will react to this increase. Yet, fertilisers are one of the most important inputs in a crop system, both in economical and environmental points of view. Hence, an effort has been made in the framework of the project to identify the marginal N, P and K fertilisers. The question thus is which N, P and K fertilisers on the market are likely to react, which producing technologies are involved and where in the world is the increased supply likely to come from? This of course depends on the location of the demand, in this case Denmark.

3.3.1 Nitrogen fertiliser

Recent consequential LCA (Nielsen et al., 2005; Dalgaard et al., 2008; Schmidt, 2007) identified calcium ammonium nitrate produced in Eastern European plants to be the less competitive supplier of mineral nitrogen (N). Two of these studies (Nielsen et al., 2005; Dalgaard et al., 2008) based their market analysis on the European market, and claimed that the least competitive fertilizer is the one affected, based on a decrease in consumption in the European market due to environmental restrictions. In contrast to this, Schmidt (2007) argues that the geographical market for fertilizer is not limited to Europe, based on the import share of the supply to EU25 in 2005, which is over 20 % for ammonia, ammonia nitrate and calcium ammonia nitrate. Therefore, Schmidt (2007) proposed to identify the marginal N based on the global market.

For all these studies, the basis for identifying the marginal N lies in historical data. Yet, LCA aims to assess the consequences (occurring in the future) of the implementation of given scenarios, so forecasts based on state-of-the-art models shall be prioritized over simple extrapolation of historical data.

Recent forecasts indicate increases of N use for both the world and Western Europe, and it is concluded that a rising trend for N fertilizer should be taken into account in determining the marginal. A rising market trend involves, based on the consequential principles presented by Weidema (2003), that the most competitive supplier is the one affected by a change in demand. It is, however, acknowledged that for N fertilisers the trend may not only be market driven and influenced by a number of factors like agri-environmental measures or other policy intervention types (e.g. CAP measures in EU countries), which are common in the agricultural sector.

Based on a further analysis of the developments in the fertilizer production (see inventory report in the WP5 annex), two N fertilizers may be distinguished as the potential marginal, depending on the market considered: urea (global market) and nitrate based fertilizers (European market) (ammonium nitrate or calcium ammonium nitrate).

It should, however, be highlighted that both urea and calcium ammonium nitrate are derived from the same substance, ammonia (NH_3), which is the richest source of N for any of the synthetic N fertilisers available (Longacre et al., 2010). A proportion of 97 % of nitrogen fertilisers are derived from ammonia (EFMA, 2004).

Today, ammonia is produced synthetically through the so-called “Haber-Bosch process”, a high-pressure catalytic process using, as N source, the N from air. The natural gas is generally the most competitive source for the needed hydrogen (H) (EFMA, 2009). While the N from air is not constrained in supply (N represents 78 % of the air composition), the supply in natural gas is subjected to constraints. Moreover, according to EFMA (2009), natural gas represents between 50 and 70 % of the total feedstock cost, meaning that the cost of natural gas is an important parameter in the price of N fertilisers. In the light of this, it appears that the ultimate product affected by a change in demand in N fertilisers (under a rising trend) is the supply for H to synthesize the ammonia necessary to produce all fertilisers. As mineral N fertilisers are, in Denmark, essentially imported, this important interaction with countries still using fossil fuel is important to keep in mind when interpreting the results of the scenarios presented in this project, i.e. demanding more N fertilizers in Denmark because renewable energy ambitions involve more import of these, and thereby more natural gas consumed somewhere else.

If natural gas is considered a constrained resource, it means that a prioritization of its various uses has to be made. If its use for the fertiliser industry is prioritized, this natural gas is not available for other competing uses, for instance CHP production, meaning that a demand for other C source materials in CHP production is created, e.g. coal, or eventually biomass in case the fertiliser comes from a region with a 100 % renewable energy system. In this specific case (displaced natural gas towards the fertiliser industry giving rise to an increase in biomass-for-energy), this is likely to involve a demand for additional fertiliser (for producing this biomass-for-energy), besides leading to arable land expansion. Such a scenario appears rather unsustainable. This also applies in the case of hydrogen being produced by biomass which is of growing interest (e.g. Kalinci et al., 2009; Balat and Kirtay, 2010). On the other hand, if natural gas is not considered a constrained resource, then the cause-effect relation is straight-forward: a rising demand for fertilizer involves a rising demand for natural gas. This in turn may interact with the energy market by a change in price that could have repercussions in substituted energy products (e.g. coal).

3.3.2 Phosphorus fertiliser

Forecasts for P demand also tend towards a global increase. Short-term projections for P_2O_5 consumption in “West and Central Europe” (FAO, 2009) indicate stability (consumption 3 M tonnes in 2008 and in 2013). The longer-term forecasts (up to 2030) made by Tenkorang and Lowenberg-DeBoer (2009), however, indicate a consumption increase in all regions considered in the model, except for Eastern Europe and former soviet countries. Tenkorang and Lowenberg-DeBoer (2009) projections for EU indicate a commercial P consumption of 3.1, 4.3 and 5.2 M megatonnes for 2005, 2015 and 2030, respectively. For the world, their forecasts indicate a commercial P consumption of 36.6, 43.8 and 52.9 M megatonnes (for 2005, 2015 and 2030, respectively). These forecasts, however, may be underestimated as they include neither the recent increases in biofuel demand nor an economic growth continuing in developing countries (particularly China and India). For EU-15, however, phosphorus consumption is forecasted to decline by 13.6 % on the short-term horizon (2008-2018) (EFMA, 2009).

FAO (2009) reports that close to 40 new monoammonium phosphate, diammonium phosphate and triple superphosphate units will be constructed during 2008-2013 in 10 different countries, and nearly half of it should be in China. Other facilities are also planned in Africa, West Asia, East Asia and Latin America. Most of these 40 new units should be diammonium phosphate units. Based on statistics from the International Fertiliser Association, diammonium phosphate is, in both “Western Europe” and the world, the P fertiliser with the greatest apparent consumption for the period 1999-2008 (IFA, 2010) (compared to monoammonium phosphate and triple superphosphate). Based on this, diammonium phosphate is considered to be the marginal P fertiliser in this study.

3.3.3 Potassium fertiliser

Long-term projections for K fertiliser consumption by Tenkorang and Lowenberg-DeBoer (2009) also indicate an increase trend, for EU and worldwide. This corresponds to a consumption of 3.2, 5.0 and 6.0 M megatonnes in 2005, 2015 and 2030, respectively, for the EU. For the world, the forecasts show a consumption of 26.6, 28.5 and 32.8 M megatonnes for 2005, 2015 and 2030, respectively.

In 2009, the demand for potash dropped to its lowest level in 30 years (Heffer and Prud’homme, 2009). However, strong demand prospects in the medium term have prompted many prospective producers to invest in potash projects and global potash capacity is forecasted to increase from 40 M tonnes K_2O in 2008 to 54 M tonnes K_2O in 2013 (FAO, 2009).

Varieties of potassium fertilisers include potassium chloride, potassium sulphate and potassium nitrate. However, potassium chloride (KCl) accounts for about 95 % of all potassium fertilisers used in agriculture, being the cheapest per tonne (Jonhston, 2003). Potassium chloride is therefore considered the marginal K fertiliser in this study.

4. Technological options for biomass conversion to energy

4.1. Anaerobic digestion

Anaerobic digestion of different agricultural residues (e.g. manure and grass) and organic waste is an established technology in Denmark (Raven and Gregersen 2007). A literature review of the efficiencies (in terms of biogas yield) related to the fermentation of selected biomass resources was done in Article 2. The focus was on the two major biomass resources suitable for biogas production, i.e. manure and grass.

With respect to grass, the yield was found in the range of about 200-1000 Nm^3/tonne DM (dry matter) depending on the water content (harvesting season) and type of grass. In fact, the DM content can vary significantly depending on type of grass as well as harvesting season (Prochnow et al. 2009). In Jungbluth et al. (2007), the DM content was assumed to be 15%. An average yield of 400 Nm^3/tonne DM was assumed in Article 2. The process selected for grass conversion to biogas was a grass refinery as described by Jungbluth et al. (2007). In this process, biogas, solid biofuels and grass proteins for animal feed were produced. The process was selected as it allowed to partly recover animal feed. It has to be noted, in fact, that a large share of the available grass in Denmark is typically used as feed for animals (Foedevareministeriet, 2008) and the use of the same biomass for energy purposes would determine cascade effects in the market (this was discussed in Article 2).

The biogas yield of manure was found in the range 19-28 Nm^3/tonne FM (fresh matter). The high water content of this substrate is a limiting factor for achieving high potential from an energy point of view. However, producing biogas from livestock slurry is a technique with high potential to reduce greenhouse gas (GHG) emissions, probably the best of all slurry management techniques for this purpose (Hamelin et al., 2010). As a versatile energy carrier with high exergy, there is a variety of fates for the produced biogas such as combined heat and power production (CHP), direct substitution of natural gas (through injection in the natural gas grid or as a source of hydrogen for the chemical industry), use as a liquid fuel for transportation, or even as a combustible for fuel cells. This makes biogas an integral part of renewable energy strategies, especially for a country with high animal density such as Denmark.

In addition to producing biogas, anaerobic digestion of animal slurries also results in the production of a digested slurry with enhanced efficiency as an organic nitrogen (N) fertilizer as a result of the increased ammonium (NH_4^+) content of the digested slurry (Birkmose and Petersen, 2004). In countries with high animal density, anaerobic digestion thus represents an interesting avenue for both slurry management and renewable energy production, given the large quantities of slurry produced.

Yet, animal slurry alone often contains too little easily degradable carbon (C) in order to ensure a methane (CH_4) yield, and consequently an energy output, sustainable from an economical point of view. For this reason, animal slurries are generally co-digested with substrates providing additional C input (e.g. organic wastes, energy crops, glycerin by-product from esterification of rapeseed, crop residues). However, the availability of such organic materials is rather limited as compared to the slurry availability and these substrates are also solicited for alternative uses (e.g. energy recovery through incineration, liquid fuels, etc.).

As an alternative to the addition of external C, the possibility to produce biogas from slurry only, using slurry separation as a pre-treatment to concentrate the degradable C content and thereby increase the CH_4 potential, has been investigated in Article 1 enclosed with this report. The consequential life cycle assessment performed in that study highlighted that this biogas production possibility yields significant environmental benefits, although those are really much dependent upon the efficiency of the separation technology used for separating the volatile solids of the slurry in the solid fraction. Nevertheless, this possibility is envisioned to represent the future for slurry biogas production in a 100% renewable energy system, given the constraints on C co-substrates.

4.2 Thermal gasification

Thermal gasification is a sub-stoichiometric combustion of the fuel. The process generates two major outputs: syngas (containing CH_4, H_2, CO and CO_2) and char. The syngas has a lower heating value (LHV) typically around 4-8 MJ/Nm^3, if air is used as oxidizing agent in the process. If steam is used, the LHV of the gas can be up to 16-20 MJ/Nm^3 (Arena et al. 2003). Char is a residue containing a high share (10-20%) of unconverted carbon. Thus, the final use of the char can be combustion (though difficult for the high content of ash), use-on-land or landfilling.

Thermal gasification was selected in Article 2 for the energy conversion of selected lignocellulosic biomasses such as wood, willow and straw. The choice of gasification was related to the need for energy carriers (i.e. gas) which could be fed directly into the existing natural gas grid. This assured flexibility for the whole energy system.

Thermal gasification was demonstrated to be particularly efficient with respect to the treatment of woody materials (e.g. woodchips, wood pellets, wood residues and straw), where the conversion of the energy in the biomass into energy in the syngas can reach efficiencies as high as 85%-95%, in accordance with the findings of (among others) Arena et al. (2010), Arnavat et al. (2010), Carpenter et al. (2010), Ptasinski (2008) and Ahrenfeldt et al. (2006). The syngas produced can be used as fuel for Solide Oxide Fuel Cell (SOFC) or Integrated Gas Combined Cycle (IGCC) plants reaching electricity and heat efficiencies of 50%-60% and 40%-30%, respectively (DEA, 2010). The biochar was assumed to be used-on-land (the influence of this assumption was assessed in the sensitivity analysis in Article 2). The environmental benefits derived from application of biochar to agricultural soil were discussed by Roberts et al. (2010) and considered in the LCA study in Article 2.

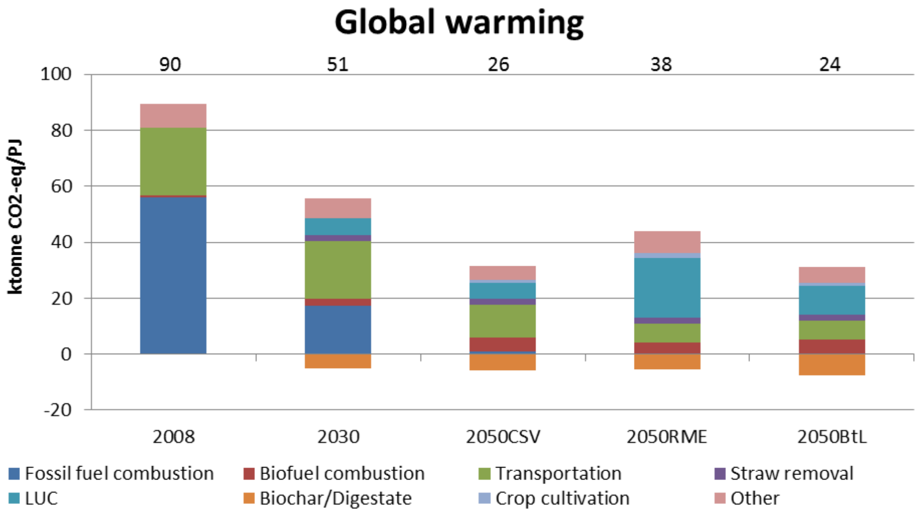
5. Life Cycle Assessment results and main related findings

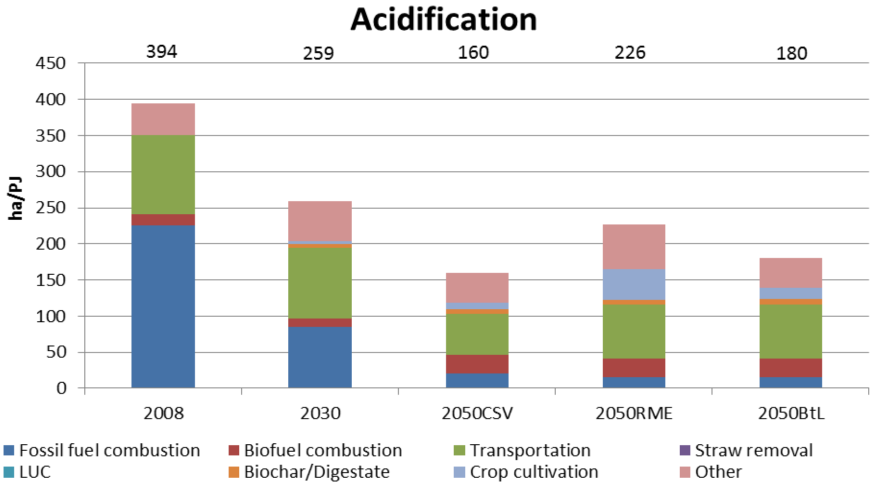
The results of the LCA were reported in Figure 2 for the selected environmental categories: global warming (GW), acidification (AC), aquatic eutrophication (AE) and land occupation (LO). The impacts were reported per unit of energy supplied by the system (e.g. ktonne CO_2-eq/PJ). The details of the investigations can be found in Article 2. In Appendix B.2, the flow charts of selected energy processes (e.g. RME production from rapeseed) were reported. Such flow charts provide an idea of the cascade effects generated by the use of biomass for energy purposes. The main findings of the LCA are summarized as follows:

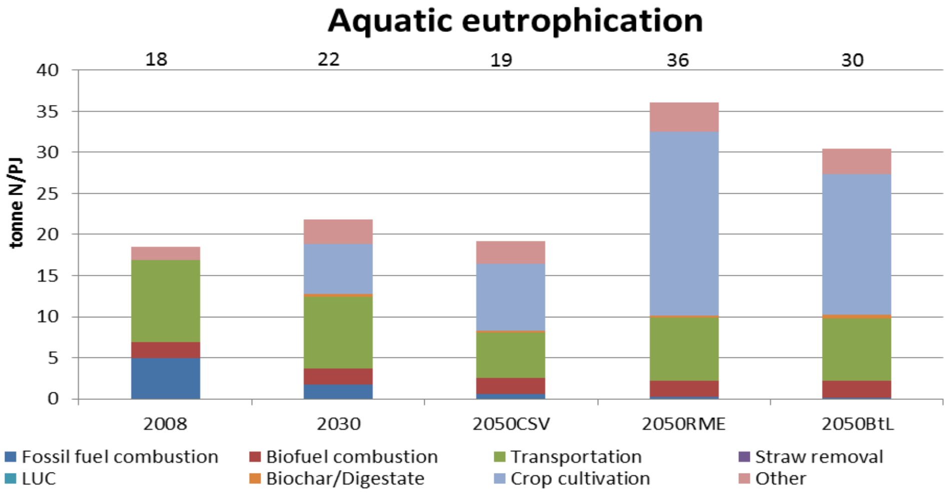
- A consistent abatement of the GHGs (about 60% - 80%, per PJ of primary energy in the system) can be achieved by implementing such energy systems. However, the residual domestic biomass resource was not sufficient to cover the energy demand. Cultivation of energy crops (e.g. willow) was thus required.
- The impacts on the category acidification followed the trend of GW, with a strong decrease of the impact per unit of energy compared to the current energy system.
- Cultivation of energy crops led to high eutrophication impacts on aquatic ecosystems. The environmental load was estimated about double as today per unit of energy supplied by the system.
- Land use change impacts (especially indirect) were characterized by a high level of uncertainty. The results underlined that high impacts related to land use changes can make the option of using fossil fuels for heavy transport favorable to the production and use of biodiesel-like fuels.

The main challenge of such future energy systems was the provision of biofuels for heavy vehicles and aviation. With respect to aviation biofuels, very few studies were found in the literature and the suggested processes were considered not mature from a technical point of view. Thus, it was assumed that aviation fuels still relied on fossil resources. With respect to terrestrial transportation, the low efficiency of second generation ethanol technologies and transesterification processes determined the need for cultivation of energy crops (e.g. rapeseed) generating significant environmental impacts on global warming (land use changes) and aquatic eutrophication (increased fertilizer use) as well as increased land occupation. The impacts associated with land use changes were in some cases off-setting the benefits associated with the biofuels. This, however, strongly depended on the assumptions regarding the magnitude of dLUC and iLUC.

As mentioned earlier, the results showed that increased use of fertilizers for energy crops cultivation could potentially lead to high impacts on aquatic eutrophication due to nutrient release to surface water. The use of land was estimated to increase compared to the current situation of about 600-2100 ha/PJ depending on the amount of energy crops cultivated (see Article 2).


Global warming
90
51
26
38
24
100
80
60
40
20
0
-20
ktonne CO2-eq/PJ
2008
2030
2050CSV
2050RME
2050BtL
Fossil fuel combustion
Biofuel combustion
Transportation
Straw removal
LUC
Biochar/Digestate
Crop cultivation
Other


Acidification
394
259
160
226
180
450
400
350
300
250
200
150
100
50
0
ha/PJ
2008
2030
2050CSV
2050RME
2050BtL
Fossil fuel combustion
Biofuel combustion
Transportation
Straw removal
LUC
Biochar/Digestate
Crop cultivation
Other


Aquatic eutrophication
18
22
19
36
30
40
35
30
25
20
15
10
5
0
tonne N/PJ
2008
2030
2050CSV
2050RME
2050BtL
Fossil fuel combustion
Biofuel combustion
Transportation
Straw removal
LUC
Biochar/Digestate
Crop cultivation
Other

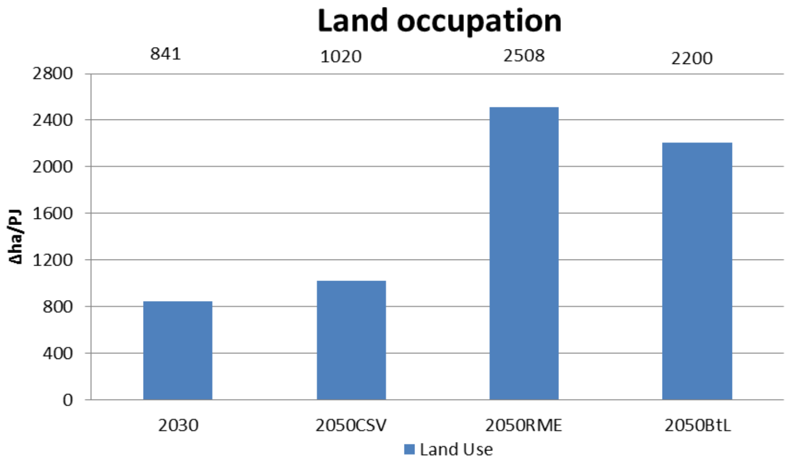


Figure 2 Result of the LCA for selected environmental categories. The values are expressed per unit of energy (PJ) supplied by the energy system

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7. List of written manuscripts

This section contains the scientific papers written in the framework of this project. Given that the two PhD students joined the project at its late stage, these papers are to be submitted in 2011. These papers are joined to this report. Changes to the versions presented here shall be expected after the revision process.

1. Hamelin L, Wesnæs M, Wenzel H, Petersen B M (2011): Environmental consequences of future biogas technologies based on separated slurry. Accepted for publication in *Environmental Science & Technology*.
2. Tonini D, Astrup T. (2011): Environmental assessment of future sustainable energy systems for Denmark. To be submitted to *Biomass & Bioenergy*.

8. List of manuscripts envisioned

Additional papers related to this project, currently under redaction or to be written in the course of 2011, are also envisioned. These are listed below, but subjected to changes:

1. Hamelin L, Jørgensen, U, Petersen B M, Olesen J E, Wenzel H (2011): Modelling the environmental consequences of direct land use changes from energy crops in Denmark: a consequential life cycle inventory. To be submitted to *Global Change Biology Bioenergy*.
2. Tonini D, Hamelin L, Wenzel H, Astrup T, (2011): Carbon flows and environmental impacts of energy and fuel production from selected perennial crops for Denmark. To be submitted to *Biomass & Bioenergy*.
3. Hamelin L, Wenzel H (2011): Modeling the indirect land use change consequences of energy crops in life cycle assessment – a life cycle inventory for Denmark. To be submitted to *International Journal of LCA*.
4. Hamelin L, Wenzel H, Jørgensen U (2011). Consequential life cycle assessment of direct land use changes from Danish energy crop systems: effect of crop type, soil, climate, residues management, initial soil carbon level and turnover time.

9. Other scientific activities related to the project

9.1 Poster presentation

1. Hamelin L, Petersen B M, Jørgensen U, Olesen J E, Wenzel H (2010): A consequential LCI for energy crops in a future Danish renewable energy system. Poster presented at the VII International Conference on Life Cycle Assessment in the agri-food sector (LCAfood 2010). Bari, Italy, September 22nd – 24th.

9.2 Oral presentations in conferences and seminars

1. Hamelin L: (2010). Life cycle assessment of biogas form separated slurry. Presented at: XVIIth World Congress of the International Commission of Agricultural and Bioengineering. Quebec city, Canada, June 13th – 17th.
2. Hamelin L: (2010). Biogas, slurry separation & acidification: a few LCA perspectives based on recent studies. Presented at: Seminar om biogas og gylleseparation. Odense, Denmark: October 13th.
3. Hamelin L: (2010). Life cycle assessment of biogas form separated slurry. Presented at: Annual 3R Research Seminar. Copenhagen, Denmark: June 3rd.

9.3 Scientific reports

1. Hamelin L, Wesnæs M, Wenzel H, Petersen B M (2010): Life cycle assessment of biogas from separated slurry, Environmental project no. 1329; Danish Ministry of the Environment, Environmental Protection Agency: Copenhagen, Denmark; http://www.mst.dk/Publikationer/Publications/2010/07/978-87-92668-03-5.htm

9.4 Miscellaneous

1. Hamelin L: (2011). Inventory report for modelling direct land use changes of perennial and annual crop in Denmark. Unpublished.
2. Tonini D, Hamelin L, Astrup T, Wenzel H (2011). Flow charts of selected biomass conversion technologies. Unpublished.

Appendix A. Written manuscripts

A.1. Article 1: Biogas from separated slurry

Hamelin L, Wesnæs M, Wenzel H, Petersen B M (2011): Environmental consequences of future biogas technologies based on separated slurry. Accepted for publication by Environmental Science & Technology.

Environmental Consequences of Future Biogas Technologies based on Separated Slurry

LORIE HAMELIN†, MARIANNE WESNÆS†, HENRIK WENZEL†, BJØRN M. PETERSEN‡*

†Institute of Chemical Engineering, Biotechnology and Environmental Technology, Faculty of Engineering, University of Southern Denmark, Campusvej 55, 5230 Odense M., Denmark

‡ Department of Agroecology and Environment, Faculty of Agricultural Sciences, Aarhus University, Blichers Allé 20, Postboks 50, 8830 Tjele, Denmark.

* Corresponding author phone: +45 20585159; fax: +45 65507354; e-mail: loha@kbm.sdu.dk (L.H)

ABSTRACT. This consequential life cycle assessment study highlights the key environmental aspects of producing biogas from separated pig and cow slurry, a relatively new but probable scenario for future biogas production, as it avoids the reliance on constrained carbon co-substrates. Three scenarios involving different slurry separation technologies have been assessed and compared to a business-as-usual reference slurry management scenario. The results show that the environmental benefits of such biogas production are highly dependent upon the efficiency of the separation technology used to concentrate the volatile solids in the solid fraction. The biogas scenario involving the most efficient separation technology resulted in a dry matter separation efficiency of 87 % and allowed a net reduction of the global warming potential of 40 %, compared to the reference slurry management. This figure comprises the whole slurry life cycle, including the flows by-passing the biogas plant. This study includes soil carbon balances and a method for quantifying the changes in yield resulting from increased nitrogen availability as well as for quantifying mineral fertilizers displacement. Soil carbon balances

showed that between 13 and 50 % less carbon ends up in the soil pool with the different biogas alternatives, as opposed to the reference slurry management.

INTRODUCTION

Making biogas from animal slurry is a priority option for reducing greenhouse gas (GHG) emissions and contributing to renewable energy supply in many countries. In Europe, an increase in slurry based biogas is envisioned as a key element in emerging renewable energy strategies, motivated by the European Union target of achieving 20% renewable energy by 2020 (1). For example, the Danish government proposed a target of using 50 % of the manure produced in Denmark for renewable energy by 2020, which would essentially be met through a strong biogas expansion (2). In Germany, over 4000 large scale biogas plants were built since the late 1990'ies. When designed and operated properly, ensuring e.g. against methane (CH_4) losses from the degassed slurry, slurry biogas has been found to be one of the most cost-effective ways of reducing GHG emissions due to simultaneous benefits of reduced CH_4 and nitrous oxide (N_2O) emissions from slurry storage and field application as well as of replaced fossil fuels from utilizing the biogas (3). The cost was found to be around 13 Euro ton^{-1} carbon dioxide equivalent (CO_2 eq.), being lower than most other measures for GHG reduction and one of the largest contribution to GHG reduction and renewable energy supply agriculture can make. There are, however, two major obstacles for a widespread implementation of slurry biogas. First, animal slurries are often too dilute, containing too little easily degradable carbon (C) for ensuring economically attractive CH_4 yields. Further, the supply of nitrogen (N) from slurry often exceeds the demand for microbial growth during the anaerobic digestion process (i.e. too low C:N ratio), leading to accumulation of ammonia (NH_3) and potentially to some inhibition of the CH_4 producing bacteria (4). These obstacles have traditionally been solved by supplementing the slurry with substrates providing additional C input. In Denmark, the strategy used so far has been to use C rich and easily degradable industrial wastes as a co-substrate. However, the availability of applicable organic residues is rather limited compared to slurry volumes. In e.g. Denmark, around 5 % of the slurry goes through a biogas plant with co-digestion of organic industrial residues, and this requires almost all suitable residues available (5). So with Denmark as an example, more than 90 % of animal slurry will need another strategy for increasing the economic feasibility of biogas. Alternative strategies, however, do exist: (i) to use energy crops as external C input, (ii) to change housing systems into systems keeping the animal urine and feces apart, thus producing a solid manure very economically attractive for biogas, (iii) to separate the slurry from existing slurry-based housing systems into a dilute and a concentrated fraction, and use this concentrated fraction as a co-substrate to raw slurry; or (iv) to accept the dilute slurry and the related low biogas yields and compensate this through bigger digesters with higher slurry retention times. All strategies have their advantages and disadvantages, economically as well as environmentally. The slurry separation strategy is the one investigated in this study.

Such a biogas production concept has been tested in pilot scale experiments (6-7), and was pointed out by the group of companies and organizations involved in the Danish Partnership for Industrial Biotechnology (8) as a promising emerging technology. The slurry separation strategy is motivated by a wish to avoid using energy crops due to the problem of competition for land with the food sector this involves (9), and by a need to increase economic feasibility compared to using the dilute raw slurry. As part of the feasibility assessment of this concept, its environmental performance is assessed by a life cycle assessment (LCA).

The aim of this study is, thus, to compare the environmental consequences of making biogas from separated slurry to a business-as-usual reference slurry management scenario, involving three alternative slurry separation and biogas scenarios with pig slurry as well as one with dairy cow slurry. The scenarios reported in this manuscript are the pig slurry scenarios, whereas the dairy cow slurry scenario is available in the Supporting Information. The study is set up in a way allowing for future comparisons with any alternative ways of slurry management.

METHODS

ENVIRONMENTAL ASSESSMENT METHOD. The analysis is performed using consequential

LCA. LCA is a standardized environmental assessment methodology (10-11) assessing the potential environmental impacts and resources used by alternative product or service systems throughout their whole life cycle. Consequential LCA compares the differences between alternatives. This implies that the processes and/or suppliers included in the model are those responding to changes in demand by corresponding changes in supply; by definition these are the marginal suppliers (12). A consequential LCA also implies that the system is expanded in order to reflect all consequences arising when choosing a given alternative to the prevailing reference, or one alternative over another. Further elaboration on the consequential LCA approach can be found in (13-14).

FUNCTIONAL UNIT. In order to make alternatives comparable, it has to be ensured that they provide the same services to society. To do this, a functional unit is defined (10-11) and all input and output flows are expressed per functional unit. In the present study, the service provided to society is the management of slurry, and the functional unit is defined as “the management of 1 ton of post-animal slurry”, i.e. slurry as freshly excreted by the animals.

SCOPE. The geographical scope of the slurry management system (e.g. housing systems, storage facilities, legislation for fertilization, etc.) is Denmark. Any systems affected outside Denmark, e.g. fertilizers production, are obviously also included, in accordance with consequential LCA principles. The technological scope for biogas is the best technologies available in Denmark, which are further detailed in the Supporting Information. The temporal scope is 30 years, based on the life time of the technologies studied.

REFERENCE SCENARIO. The study assesses the environmental consequences of producing biogas from separated slurry and compares it to a reference slurry management scenario (REF-pig), i.e. using pig slurry as a fertilizer for crop production without prior treatment (Figure 1). This reference scenario needs to be defined in terms of housing, storage, transport distances, field spreading, soil types as well as crop rotations, for assessing the reference nutrients uptake. A complete description of these preconditions is presented in the Supporting Information.

Because slurry composition is the basis for assessing the subsequent emission flows and performing mass balances, a reference slurry has also been defined. This reference slurry was determined based on the Danish normative system for assessing slurry composition (15-16). Core parameters of the reference slurry composition considered are presented in Table 1, for the three main life cycle stages of slurry, i.e. post-animal, post-housing (as it leaves the in-house storage) and post-storage (as it leaves the outdoor storage). Additional details about the reference slurry are available in the Supporting Information.

ALTERNATIVE BIOGAS SCENARIOS. Three scenarios are assessed (P1, P2, P3), each considering different slurry separation technologies to obtain the solid fraction (SF) input for biogas production, which is to be digested together with raw un-separated slurry. In all alternatives, the produced biogas is used for combined heat and power production (CHP), which consequently displaces the marginal heat and electricity sources in the adjoining energy systems, as further detailed in a later section (identification of marginals). Similarly, slurry fractions are used as organic fertilizers, which avoid the use of corresponding marginal mineral fertilizers.

The process flow diagrams of the three alternatives are illustrated in Figure 1. In this figure, all involved flows are related to the functional unit, i.e. the excreted ton of slurry. In Table 1, the mass and nutrients share of the raw slurry transferred to the SF, referred to as separation efficiency, is presented for all separation technologies considered. Table 1 also shows the composition of all processed slurries involved in each scenario. These can be related to the functional unit through the flows presented in Figure 1. The complete mass balances performed to sustain these compositions are presented in the Supporting Information.

ALTERNATIVE P1: DECANTER CENTRIFUGE WITH POLYACRYLAMIDE. The separation process considered in P1 is a conventional centrifugal separation technology, which is combined with the addition of 0.90 kg of cationic polymer, namely polyacrylamide (PAM), per ton of slurry input to the centrifuge. The liquid fraction (LF) obtained from the separation process is stored and used as a fertilizer, while the degassed slurry (deg. slurry) resulting from the anaerobic digestion is again separated with a centrifuge, but without polymer addition. This second separation is justified by the potential for an enhanced phosphorus (P) management, given the richness of the P content in the degassed slurry, a consequence of the high separation efficiency of the first separation (Table 1). The resulting degassed liquid (deg. LF) and solid (deg. SF) fractions are then stored and used on the field as fertilizers. Because the plant availability of slurry N is increased by the anaerobic digestion process (17), an increased plant yield was also modeled, as further detailed.

ALTERNATIVE P2: SCREW PRESS. In alternative P2, the SF is produced from a mechanical screw press technology, and as in alternative P1, the LF is stored and used as a fertilizer. The degassed slurry is not separated as its P content is not high enough to justify a second separation. It is consequently simply stored and used directly as a fertilizer.

ALTERNATIVE P3: SCREW PRESS AND PELLETS PRODUCTION. Alternative P3 is identical to alternative P2, except that the produced SF is not directly used as an input for biogas, but as an input for producing fibre pellets (FP). This process consists of drying the SF in a tumble dryer and subsequently pressing it to form pellets with a dry matter (DM) content of 89 %, so transportation costs are reduced. It is these pellets that are used as an input for biogas production. However, 40 % of the produced pellets are combusted for producing the heat required for the process itself, and thus not available for biogas production. Ashes from burned pellets are used as potassium (K) and P fertilizer.

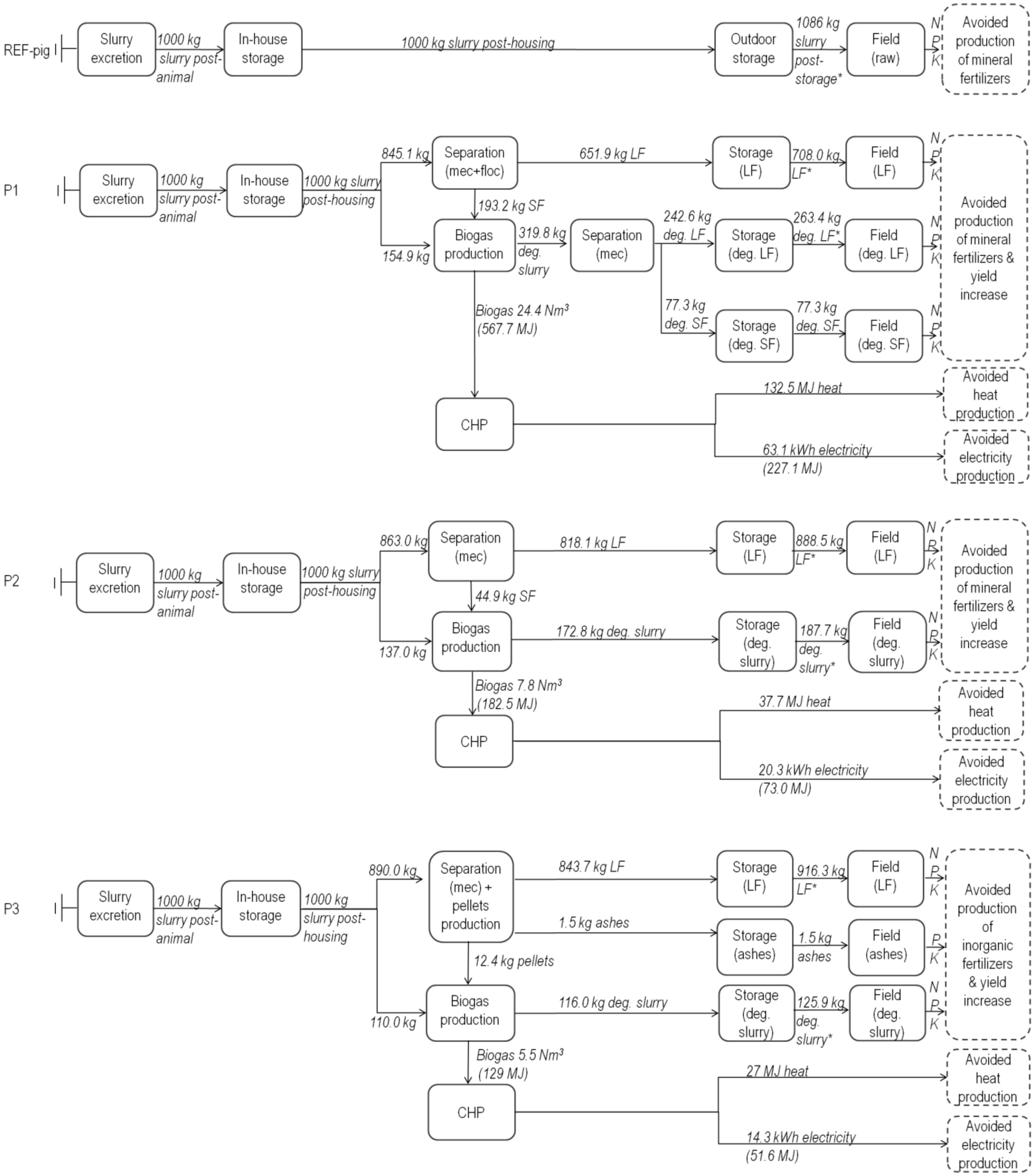

Figure 1. Process flow diagrams of the alternatives compared for pig slurry: (REF-pig) Reference system, (P1) Alternative P1: decanter centrifuge with polyacrylamide polymer, (P2) Alternative P2: screw press, (P3) Alternative P3: screw press and pellets fabrication. The dotted lines indicate avoided 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.

Table 1. Separation efficiencies of the technologies considered and composition of the reference slurry and of the different slurry fractions[a]

Parameter[b]	Mass	Total N	P	K	DM	C
Reference slurry composition (raw slurry)						
REF-pig, post-animal (kg ton^{-1} slurry post-animal)		6.60	1.1	2.9	77	37
REF-pig, post-housing (kg ton^{-1} slurry post-housing)[c]		5.48	1.1	2.9	70	33
REF-pig, post-storage (kg ton^{-1} slurry post-storage)		4.80	1.0	2.6	61	29
Separation efficiencies (% in solid fraction)						
Alternative P1 (decanter centrifuge with PAM[d])	22.9	41.9	90	14.2	87.2	87.2
Alternative P1 (decanter centrifuge, second separation)	24.2	21.2	66.2	9.7	60.9	60.9
Alternative P2 and P3 (screw press)	5.2	6.8	9.1	2.9	29.6	29.6
Solid fractions and pellets composition, prior to input for biogas (kg ton^{-1} solid fraction or pellets)						
SF, Alternative P1 (decanter centrifuge with PAM[d])		10.0	4.5	1.8	266	127
SF, Alternative P2 (screw press)		7.2	2.0	1.6	397	190
FP, Alternative P3 (screw press and pellets production[e])		11.8	4.4	3.6	889	425
Liquid fractions composition, prior to storage (kg ton^{-1} liquid fraction)						
LF, Alternative P1 (decanter centrifuge with PAM[d])		4.13	0.15	3.2	12	6
LF, Alternatives P2 and P3 (screw press)		5.4	1.1	2.9	52	25
Degassed solid fraction composition, prior to storage (kg ton^{-1} degassed solid fraction)						
Deg. SF, Alternative P1 (decanter centrifuge)		7.65	8.9	1.0	267	130
Degassed liquid fraction composition, prior to storage (kg ton^{-1} degassed liquid fraction)						
Deg. LF, Alternative P1 (decanter centrifuge)		9.06	1.4	2.9	55	27
Degassed slurries composition, prior to storage (kg ton^{-1} degassed slurry)						
Deg. slurry, Alternative P2		6.2	1.4	2.7	106	51
Deg. slurry, Alternative P3		6.5	1.5	3.1	106	51

[a] The aim of this table is to present the core composition of the different slurry fractions involved in the reference and alternative scenarios, not to present a mass balance. Mass balances behind the values shown here as well as values for additional parameters are presented in the Supporting Information.

[b]The volatile solids (VS) are not presented, but for the reference slurry as well as all SF, VS have been assumed to constitute 80 % of the DM content, based on (18). [c]The REF-pig post-housing slurry is the slurry going through separation. [d] Polyacrylamide polymer. [e]A mass loss of 28.8 kg as well as a N loss of 0.1 kg are assumed to occur during the drying process, based on data from the technology provider.

DATA SOURCE. Data for foreground processes (e.g. all data related to slurry management) were based on original data from suppliers of the relevant technologies and from various Danish studies, but also on data obtained from dynamic modeling (e.g. for soil C changes), from internationally recognized methodologies or guidelines (e.g. IPCC) and in some cases on data from other European studies. Data for background processes (e.g. those related to energy systems and fertilizers) were obtained from the Ecoinvent 2007 v. 2.0 database (19). In case of lack of data, estimates have been made rather than leaving gaps. All data source and estimates are documented in the Supporting Information.

BIOGAS. The biogas produced is considered to be composed of 65 % CH_4 and 35 % carbon dioxide (CO_2), with a lower heating value of 23.26 MJ Nm^{-3}. The CH_4 yield of the raw pig slurry and of the solid fraction obtained from alternative P1 is 319 Nm^3 CH_4 ton^{-1} VS. The solid fraction and fibre pellets obtained in alternatives P2 and P3, respectively, have a CH_4 yield of 187 Nm^3 CH_4 ton^{-1} VS. These CH_4 yields are based on original data from Danish biogas plants (20). 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, reflecting practice of state-of-the-art operation of biogas plants. This procedure is detailed in the Supporting Information, and resulted in an input of raw slurry of 44.5 %, 75.3 % and 89.9 % by total mass, for alternatives P1, P2 and P3, respectively, the remaining representing the share of the solid fraction (or fibre pellets) input. Efficiencies of 46 % for heat and 40 % for electricity (21) are considered for the biogas engine. The internal electricity consumption is assumed to correspond to 5 % of the net electricity production, based on original data from Danish biogas plants. The internal heat consumption is calculated considering that the mixture is heated from 8 to 37 °C. Complete details regarding the energy balances for the different biogas produced in each alternative are available in the Supporting Information.

IDENTIFICATION OF MARGINALS. The two main marginals to identify in this study are the (avoided) mineral fertilizers and energy (electricity and heat). Based on medium and long term forecasts (22-23), an increase in N, P and K consumption is envisioned. An analysis of the consumption pattern for the last 10 years (24-26), as well as of the planned capacities to be installed (27), leaded to identify ammonium nitrate, diammonium phosphate and potassium chloride as the marginal fertilizers. For electricity, a mixed electricity marginal based on a comprehensive energy system analysis for the Danish energy system has been used. The complex electricity marginal selected consists of 1 % wind, 48 % coal and 51 % natural gas, which is adapted from the simulation performed by (28). For heat, which, as opposed to electricity, is traded on a local market, a marginal consisting of 100 % coal was assumed. The importance of this assumption is tested as a sensitivity analysis, as further detailed in a later section (sensitivity analysis). Moreover, it was assumed that only 60 % of the surplus from the biogas plant (i.e. what remains after using the heat for the process itself) is used, in order to reflect the seasonal variations in the demand for heat in Denmark. Additional details on how the marginals were identified are available in the Supporting Information.

AVOIDED PRODUCTION OF MINERAL FERTILIZERS. The use of slurry and of the different processed slurry fractions as fertilizers leads to an avoided production of mineral N, P and K (Figure 1). For N, the modelling is based on the substitution values governed by the Danish regulation (29) and on the Danish normative system for assessing slurry composition (16), as the fertilizers accounts of farmers are typically based on these rather than on exact measurements. For example, the regulation considers an efficiency of 75 % for raw pig slurry (i.e. 100 kg slurry-N substitutes 75 kg mineral N), and this is to be applied not on the actual N content of the slurry but on the N content specified by the Danish normative system , which was 5.00 kg N ton^{-1} slurry post-storage in 2008 (16). The amount of mineral N avoided is thus 3.75 kg N ton^{-1} slurry post-storage for the reference scenario as well as for alternatives P1 and P2, which corresponds, based on the flows from Figure 1, to 4.07 kg N per functional unit (ton slurry post-animal). This reflects how much less mineral N is applied per ton of slurry used as a fertilizer.

For alternative P3, it is slightly more, i.e. 4.09 kg N per functional unit, as the regulation specifies a substitution value of 85 % for the liquid portion associated to the part of the solid fraction that is combusted. These calculations are performed in (30) and further explained in the Supporting Information.

The P and K use is not correspondingly limited by Danish legislation. For these, the avoided amount of mineral fertilizer is based on the ratio between the reference crop requirements in these nutrients and the content of P and K in the slurry applied. The reference crop rotation defined in this study has an annual average requirement of 21.5 kg P ha^{-1} and 64 kg K ha^{-1}, based on the national guidelines for fertilization (31). The slurry contains 26.50 kg P ha^{-1} and 66.25 kg K ha^{-1}, as detailed in the Supporting Information. Therefore, only 81 % of the applied slurry P replaces mineral P fertilizer, the rest is simply an excess that would not have been applied otherwise, and part of this excess is estimated to reach aquatic recipients (modelling details are presented in the Supporting Information). For K, it is 97 % of the applied slurry K that replaces mineral K fertilizer. For alternative P1, the P is not applied in excess since the degassed SF (where the majority of the P ends up) is assumed to be applied to a field deficient in P.

YIELD INCREASE. As a result of anaerobic digestion, the shift towards more ammonium nitrogen (NH_4-N) in the digested effluent leads to a higher N uptake by the crops, as NH_4-N is more readily available to the plants than organic N (17). In order to reflect this, the increase in crop yield induced by the use of such more efficient organic fertilizer, compared to the reference slurry, was modeled. First, the difference between the harvested N from the crop rotation (i.e. after gaseous and leaching losses) in an alternative scenario and the N harvested from the crop rotation in the reference scenario was calculated. This difference in harvested N was then translated into a response in extra wheat, assuming a response of 9.0 kg extra wheat grain per kg N surplus (30). Wheat was chosen to illustrate the response in terms of increased yield as it is the highest yielding cereal in Denmark, so the results should be seen as higher end-of-interval values. An increased wheat yield means that the production of this extra wheat does not have to be produced somewhere else in Denmark and can consequently be deducted from the system. It is acknowledged that this approach is not fully in accordance with consequential LCA as the actual consequence of higher crop yield in Denmark is more likely to be that somewhere in the world, the least competitive(s) crop supplier(s) will be taken out of production, fully or partly. The applied approach shall therefore only be seen as a rough attempt to reflect the magnitude of the environmental impacts of increased yields from using a more efficient organic N fertilizer.

IMPACT ASSESSMENT. The impact assessment was performed with the EDIP methodology (32) and further updates of the method (33-35). The impact categories considered are those judged the most susceptible to be affected by slurry management, namely global warming (over a 100 years horizon), acidification, aquatic eutrophication (distinguishing between N and P being the limiting nutrient for growth) and photochemical ozone formation. To this, the impact category “respiratory inorganics”, which reflects the emission of particulate matters, has been added, based on the Impact 2002+ method (36).

SENSITIVITY ANALYSIS. Various sensitivity analyses were performed on one alternative (P1) in order to highlight the importance of some of the most sensitive assumptions and methodological choices. This includes soil types (clay instead of sand), time horizon for C turnover in the soil (10 years instead of 100 years), a different electricity marginal (100 % coal instead of the mix electricity marginal), a different heat marginal (natural gas instead of coal) as well as a different use of the biogas (injected into the natural gas grid instead of CHP).

RESULTS

IMPACT ASSESSMENT RESULTS. For all impact categories assessed, all biogas alternatives allowed for a net impact lower or equal to what is obtained with the reference system. This is illustrated in Figure 2, which presents the results breakdown by processes. Further, for each of the significant processes, the specific substances contributing to the different impact categories are highlighted.

The net impact for a given alternative is obtained by subtracting the avoided impacts (i.e. the negative values on the graphs shown in Figure 2) from the induced impacts (positive values). There is a benefit when the net impact of a given alternative is lower than the net impact of the reference scenario. Net impacts are presented for global warming, as this is an impact of high relevance for policy making, and as it is the impact where the greatest differences with the reference scenario are obtained.

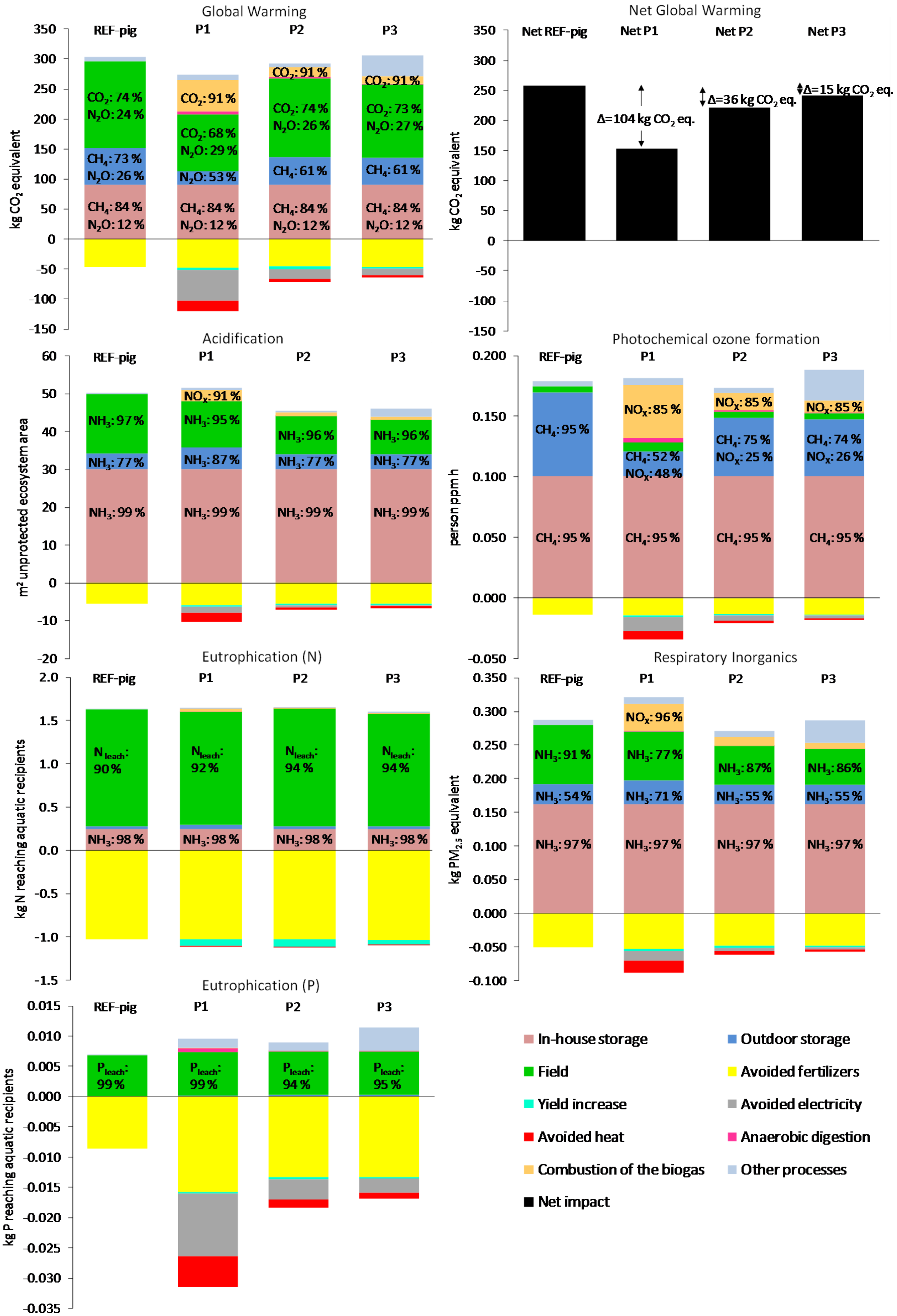
Global Warming
kg CO$_2$ equivalent
REF-pig
P1
P2
P3
CO$_2$: 74 %
N$_2$O: 24 %
CH$_4$: 73 %
N$_2$O: 26 %
CH$_4$: 84 %
N$_2$O: 12 %
CO$_2$: 91 %
CO$_2$: 68 %
N$_2$O: 29 %
N$_2$O: 53 %
CO$_2$: 74 %
N$_2$O: 26 %
CH$_4$: 61 %
CO$_2$: 73 %
N$_2$O: 27 %
Net Global Warming
Net REF-pig
Net P1
Net P2
Net P3
Δ=104 kg CO$_2$ eq.
Δ=36 kg CO$_2$ eq.
Δ=15 kg CO$_2$ eq.
Acidification
m^2 unprotected ecosystem area
NO$_x$: 91 %
NH$_3$: 97 %
NH$_3$: 95 %
NH$_3$: 96 %
NH$_3$: 77 %
NH$_3$: 87 %
NH$_3$: 99 %
Photochemical ozone formation
person ppm h
CH$_4$: 95 %
NO$_x$: 85 %
CH$_4$: 52 %
NO$_x$: 48 %
CH$_4$: 75 %
NO$_x$: 25 %
CH$_4$: 74 %
NO$_x$: 26 %
Eutrophication (N)
kg N reaching aquatic recipients
N$_{leach}$: 90 %
N$_{leach}$: 92 %
N$_{leach}$: 94 %
NH$_3$: 98 %
Respiratory Inorganics
kg PM$_{2.5}$ equivalent
NO$_x$: 96 %
NH$_3$: 91 %
NH$_3$: 77 %
NH$_3$: 87%
NH$_3$: 86%
NH$_3$: 54 %
NH$_3$: 71 %
NH$_3$: 55 %
NH$_3$: 97 %
Eutrophication (P)
kg P reaching aquatic recipients
P$_{leach}$: 99 %
P$_{leach}$: 94 %
P$_{leach}$: 95 %
In-house storage
Outdoor storage
Field
Avoided fertilizers
Yield increase
Avoided electricity
Avoided heat
Anaerobic digestion
Combustion of the biogas
Other processes
Net impact

Figure 2. Breakdown of impact assessment results for all impacts and alternatives assessed

CARBON STORED IN THE SOIL. Figure 2 illustrates the importance of biogenic CO_2 for the global warming potential of the field processes. This represents the C from the applied slurry that does not end up in the soil C pool, and was modeled with the 3-pooled dynamic soil model C-TOOL (37-38). Table 2 presents, for all alternatives, the biogenic C fate of all slurry and slurry fractions applied to the field. For the biogas scenarios, between 13 % and 50 % less C ends up in the soil pool, as opposed to the reference scenario.

Table 2. Balance for carbon stored in the soil for all assessed systems[a]

	REF-pig	P1	P2	P3
C added with slurry (kg ton^{-1} slurry post-animal)	31.71	19.45	27.62	25.41
C lost as CO_2 (field) (kg ton^{-1} slurry post-animal)	-30.68	-18.55	-26.98	-24.90
C stored in the soil (kg ton^{-1} slurry post-animal)	1.03	0.90	0.64	0.51
Net CO_2-C “stored”[b] (kg ton^{-1} slurry post-animal)	3.77	3.30	2.35	1.87

[a] The repartition between the C ending up as emitted CO_2-C and as sequestrated in the soil is based on a 100 years time horizon. [b] This is the C stored in the soil, expressed in CO_2 through the molecular weight ratios. It does not represent a sequestration of CO_2 (it is C that is sequestrated).

DISCUSSION

Figure 2 shows that producing biogas from separated slurry does allow for net environmental benefits compared to the reference slurry management alternative, for the chosen environmental impact categories. However, as it can be visualized for the global warming impact, alternative P1 allowed much greater net benefits compared to the reference system, than did alternatives P2 and P3. This is also true for the other impact categories, but in smaller magnitudes. Alternative P1 involved a separation technology (i.e. decanter centrifuge with PAM) with a much higher efficiency for DM separation (87 % compared to 30 % for alternatives P2 and P3, as shown in Table 1), and consequently concentrated better the VS in the solid fraction. This means that more of the easily degradable VS (the degradation of which produces CH_4) ended up in the anaerobic digester (i.e 50, 22 and 15 kg VS per ton slurry post-animal for alternative P1, P2 and P3, respectively), and consequently less were available for emissions to atmosphere during outdoor storage and field application (for both liquid and degassed fractions). A higher concentration of VS in the input for biogas production per ton of slurry excreted also means a higher CHP production and thereby a greater displacement of marginal energy. In a nutshell, these results indicate that the environmental benefits of the biogas production concept based on separated slurry are highly dependent upon the efficiency of the separation technology used to concentrate the volatile solids in the solid fraction.

The net figure for global warming presented in Figure 2 differs from figures typically found in earlier studies (e.g. 39), where the net contribution from biogas alternatives is practically zero, once the displaced energy is subtracted. This is because the present study considers the whole slurry flow; it starts at excretion and includes the slurry flow that by-passes the biogas plant as well as the in-house slurry storage. In fact, the processes related to the management of the liquid resulting from the first separation (i.e. outdoor storage and field application) represent 16 %, 47 %, and 46 % of the GHG emissions (as CO_2 eq.) for alternatives P1, P2, and P3, respectively. Similarly, in-house slurry storage accounted for between 30 and 33 % of the GHG emissions, for the reference and the biogas alternatives. When leaving out both this by-passed liquid fraction and the in-house storage and expressing results per slurry input to the anaerobic digester (as in earlier studies) instead of per ton post-animal slurry, our study would find close to 100 % reduction of global warming potential compared to the reference slurry management scenario. But this comparison would make no sense, as the biogas concept assessed in this study has for consequence the production of a liquid fraction that must be dealt with, which somehow represents a limit to the environmental benefits that can be obtained from this biogas production concept. On the other hand, the in-house slurry storage could have been left out of the assessment as it is not influenced by the biogas production, but it was considered relevant to include it in the perspective of broadening the study to other slurry management techniques. Moreover, results highlighted that it is an important contributor to most of the impact categories assessed (Figure 2). This is due to two substances: CH_4 and NH_3. High CH_4 emissions were expected from this process, as the anaerobic conditions for slurry below animal floors favor CH_4 formation (40). Yet, the important magnitude of

CH_4 emissions in absolute terms may be due to a conservative methodological choice, as further discussed in the Supporting Information. Ammonia emissions from in-house slurry storage has also been identified as an environmental hot spot in previous studies (e.g. 41) and mitigation measures to reduce NH_3 emissions from housing units have been the object of several studies (e.g. 42-44). Technologies allowing to reduce both CH_4 and NH_3 emissions from in-house slurry storage, e.g. slurry acidification, thus represent a clear opportunity for improving the environmental performance of slurry management.

Using the slurry as an organic fertilizer instead of mineral fertilizers is rather significant for most impact categories. Avoiding the production of marginal heat and electricity also allows significant gains, especially for global warming, while the benefit from the increased yield resulting from the use of the digested slurry appears rather negligible. At the light of this result, a more sophisticated approach to identify the exact markets reacting to an increased wheat production from Denmark due to this yield effect was not judged necessary.

None of the sensitivity analyses performed resulted in a change of the tendencies presented in Figure 2, only in changes of the magnitude of the gains obtained by the biogas production. The impact category “aquatic N-eutrophication” is the most sensitive to a change of soil (from sand to clay) and of the time horizon for C turnover (from 100 years to 10 years). Injecting the biogas in the natural gas grid instead of producing CHP led to important decreases of the differences between the reference and the biogas scenarios (e.g. 29 % decrease for the global warming impact), indicating that using the biogas directly yields more environmental benefits than upgrading it to replace natural gas. This finding is in agreement with (45). Changing the source of marginal energy (electricity and/or heat) changed the differences between the reference and the biogas scenario by no more than 5 % for all impact categories.

An important limit of the study relates to the lack of information as regarding the fate of easily and slowly degradable VS following the separation of raw slurry, i.e. how much of the easily degradable VS end up in the liquid and in the solid fractions. In this study, it has been assumed that all the VS ending up in each fraction are easily degradable, which is obviously incorrect, but this was judged as the best compromise under the current status of data availability.

Some obvious environmental implications of the systems studied, like the emission of odours or the fate of cationic PAM (used in alternative P1) could not be reflected in the present study due to a lack of data as well as to limitations of the LCA methodology to include the former in the impact assessment. Based on evidences from several studies on cationic PAM, which are summarized in the Supporting Information, it was considered that it simply accumulates in the environment. However, it has not been possible to reflect the consequences (e.g. toxicity) of this. Also not reflected in this LCA are the long-term consequences of reduced soil carbon, a drawback of the biogas alternatives compared to the reference slurry management (Table 2).

Finally, the potential for expanding pig or cow production in Denmark as a result of the introduction of separation technologies on farms has not been included. In fact, the Danish law (46) allows the farmers introducing efficient separation technologies on their farms to increase their production in terms of number of animals per area of land. This increased supply of milk and meat from Denmark would have consequences in Denmark and potentially beyond, through interacting with other suppliers for these products on the world market. The overall resulting effect from this is not straightforward, and it would certainly be worth to estimate it before implementing this biogas technique (i.e. from separated slurry) on a large scale in Denmark.

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SUPPORTING INFORMATION AVAILABLE. Scope, functional unit, pre-conditions for slurry management, technologies description, biogas alternative from separated cow slurry, life cycle inventory methodology, slurry composition, mass balances, energy balance, identification of marginals, avoided mineral fertilizers, cationic polyacrylamide.

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Supporting Information for:

Environmental Consequences of Future Biogas Technologies Based on Separated Slurry

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

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 (CH_4) 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, it 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 (kg ha^{-1} 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 (kg ha^{-1} 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	Pig	6.60	5.48	4.80	N post-animal from (14). Losses considered (during housing and during storage): NH_3, N_2O, 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].
(kg ton^{-1})	Cow	6.87	6.34	5.79	
P	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].
(kg ton^{-1})	Cow	1.02	1.03	0.98	
K	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].
(kg ton^{-1})	Cow	5.81	5.90	5.65	
DM	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).
(kg ton^{-1})	Cow	125.7	113.2	103.0	
VS	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).
(kg ton^{-1})	Cow	104.2	91.7	82.4	
C	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).
(kg ton^{-1})	Cow	55.2	49.7	45.2	
Cu	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.
(g ton^{-1})	Cow	12.1	12.1	11.6	
Zn	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.
(g ton^{-1})	Cow	23.4	23.4	22.4	

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[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_2O-N	0.002 kg N_2O-N per kg N post-animal (16)
N_2-N	Assumption that N_2-N = N_2O-N * 3 (based on data from Dämmgen and Hutchings, (17))
NO-N	Assumption that N_2-N = N_2O-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_2O-N	0.005 kg N_2O-N per kg N post-animal (16)
N_2-N	Assumption that N_2-N = N_2O-N * 3 (based on data from Dämmgen and Hutchings, (17))
NO-N	Assumption that N_2-N = N_2O-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), cooper (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	*Mass balance: amount transferred to the solid fraction (SF)*	*Mass balance: amount transferred to the liquid fraction (LF)*	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. 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	*Mass balance: amount transferred to the solid fraction (SF)*	*Mass balance: amount transferred to the liquid fraction (LF)*	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	4.6	*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)

	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]
Unit	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	*Mass balance: amount transferred to the solid fraction (SF)*	*Mass balance: amount transferred to the liquid fraction (LF)*	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	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.

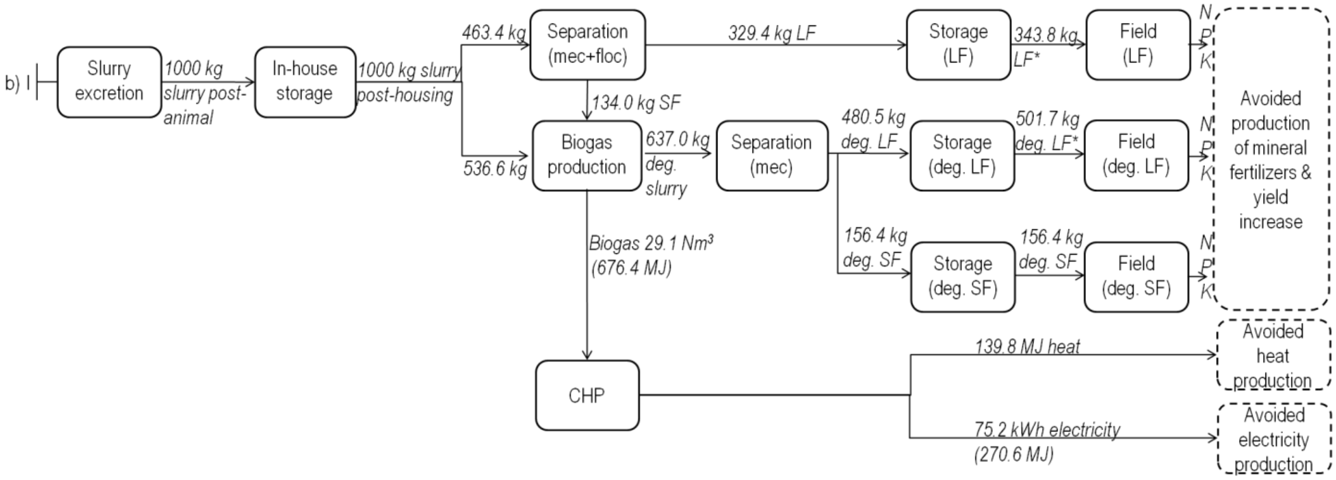

Figure S1. Process flow diagrams of the alternatives compared for dairy cow slurry management: (a) Reference system (REF-cow), (b) Alternative C1. The dotted lines indicate avoided processes. Flows marked with * include the addition of rain water. The diagrams are simplified and only include the main 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_2O_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_2O.	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 (CH_4), carbon dioxide (CO_2), ammonia (NH_3), dinitrogen monoxide (N_2O), nitrogen (N_2), 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
Emission to air	
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_2O-N (direct)	Based on IPCC guidelines (16), 0.002 kg N_2O-N are emitted per kg of N in post-animal slurry. This stands for both pig and dairy cow slurry.
N_2O-N (indirect, from NH_3 and NO_x)	Based on IPCC guidelines (16), 0.01 kg N_2O-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_2O-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_2O-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.
Discharges to soil and water	
	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 CH_4 emissions from in-house slurry storage have been slightly overestimated. The methodology used to estimate the emissions of CH_4 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 CH_4 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 Arrehenius 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_2O-N (direct)	Emission of 0.005 kg N_2O-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_2O-N (indirect, from NH_3 and NOx)	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 CO_2 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 CO_2 emissions from storage. Thus, biogenic CO_2 emissions have been estimated as a function of biogenic CH_4 releases. The ratio between CO_2 and CH_4 emitted during anaerobic degradation is estimated based on the Buswell equation (37), as presented in Equation S1:

$$C_nH_aO_b + \left(n - \frac{a}{4} - \frac{b}{2}\right) H_2O \longrightarrow \left(\frac{n}{2} + \frac{a}{8} - \frac{b}{4}\right) CH_4 + \left(\frac{n}{2} - \frac{a}{8} + \frac{b}{4}\right) CO_2 \qquad \text{Eq S1.}$$

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 (%)
VS easily degradable			
VS lipid	$C_{57}H_{104}O_6$	16.2	7.7
VS protein	$C_5H_7O_2N$	27.0	16.8
VS Volatile fatty acids (VFA)	$C_2H_4O_2$	8.5	4.0
VS carbohydrates easily degradable	$C_6H_{10}O_5$	27.1	41.5
VS slowly degradable			
VS carbohydrates slowly degradable	$C_6H_{10}O_5$	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 CO_2 and CH_4 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 CH_4 and CO_2 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
Emission to air					
CH_4	Assumed negligible, based on field experiments results (39-40).				
N_2O-N (direct)	Emission of 0.01 kg N_2O-N per kg N in slurry post-storage, based on IPCC guidelines (41).				Emission of N_2O-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_2O-N (indirect, from NH_3 and NO_x)	Same methodology as described in Table S8.				
N_2O-N (indirect, from N leaching)	Based on IPCC guidelines (41), 0.0075 kg N_2O-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_2O-N emissions.				
N_2-N	Estimated from SimDen model ratios between N_2-N and N_2O-N of 3:1, for sandy soils (46).				

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[a] Crop rotation is as described in section 3. NH_4-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. NH_4-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
Discharges to soil and water					
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 slurry 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 % CH_4 and 35 % CO_2. This composition implicitly assumes that other gases (e.g. N_2, O_2, H_2S, H_2O, CO, H_2), which altogether generally account for less than 1 % of the biogas composition, can be neglected.

Based on these proportions of CH_4 and CO_2 in the biogas and considering a CH_4 density of 0.717 kg Nm^{-3} and a CO_2 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 CH_4 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 m^3 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 obtained 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:

$$\mathbf{0.1} = \frac{\left(\left(\mathbf{W_{raw}} \times \mathbf{DM_{raw}}\right) - \mathbf{VS_{deg,raw}}\right) + \left(\left(\mathbf{W_{SF}} \times \mathbf{DM_{SF}}\right) - \mathbf{VS_{deg,SF}}\right)}{\left(\mathbf{W_{raw}} - \mathbf{W_{biogas,raw}}\right) + \left(\mathbf{W_{SF}} - \mathbf{W_{biogas,SF}}\right)} \qquad \text{Eq S2.}$$

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

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

$$\mathbf{VS_{deg,fract}} = \left(\mathbf{W_{fract}} \times \mathbf{DM_{fract}}\right) \times \mathbf{80\%} \times \mathbf{DR_{fract}} \qquad \text{Eq S4}$$

Where $VS_{deg,fract}$ 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	$W_{raw}*0.07*0.8 = 0.056*W_{raw}$	60	$0.03*W_{raw}$
Raw cow slurry	11.3	$W_{raw}*0.113*0.8 = 0.09*W_{raw}$	46.7	$0.04*W_{raw}$
SF, Alternative P1	26.6	$W_{SF}*0.266*0.8 = 0.213*W_{SF}$	60	$0.13*W_{SF}$
SF, Alternative C1	31.0	$W_{SF}*0.31*0.8 = 0.248*W_{SF}$	46.7	$0.11*W_{SF}$
SF, Alternative P2	39.7	$W_{SF}*0.397*0.8 = 0.318*W_{SF}$	37.8	$0.12*W_{SF}$
FP, Alternative P3	88.9	$W_{SF}*0.889*0.8 = 0.711*W_{SF}$	37.8	$0.27*W_{SF}$

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 CH_4 in the biogas (0.65 Nm^3 CH_4 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 VS input (kg)	CH_4 yield for the first digestion step (Nm^3 CH_4 ton^{-1} VS)	Calculation	Biogas weight (kg)
Raw pig slurry	$0.056*W_{raw}$	290	$(0.056*W_{raw}*290*1.158)/(0.65*1000)$	$0.0289*W_{raw}$
Raw cow slurry	$0.09*W_{raw}$	210	$(0.09*W_{raw}*210*1.158)/(0.65*1000)$	$0.0337*W_{raw}$
SF, Alternative P1	$0.213*W_{SF}$	290	$(0.213*W_{SF}*290*1.158)/(0.65*1000)$	$0.110*W_{SF}$
SF, Alternative C1	$0.248*W_{SF}$	210	$(0.248*W_{SF}*210*1.158)/(0.65*1000)$	$0.093*W_{SF}$
SF, Alternative P2	$0.318*W_{SF}$	170	$(0.318*W_{SF}*170*1.158)/(0.65*1000)$	$0.096*W_{SF}$
FP, Alternative P3	$0.711*W_{SF}$	170	$(0.711*W_{SF}*170*1.158)/(0.65*1000)$	$0.215*W_{SF}$

Using Equation S3 and expressing W_{raw} as $1000-W_{SF}$, 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
Raw slurry				
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*69.7*0.80*319)/ (0.65*1000*1000)	(753*69.7*0.80*319)/ (0.65*1000*1000)	(899*69.7*0.80*319)/ (0.65*1000*1000)	(800*113.2*0.80*231)/ (0.65*1000*1000)
Biogas produced (Nm^3)	12.2	20.6	24.6	25.7
Solid fraction				
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*265.9*0.80*319)/ (0.65*1000*1000)	(247*396.9*0.80*187)/ (0.65*1000*1000)	(101*889.3*0.80*187)/ (0.65*1000*1000)	(200*310.0*0.80*231)/ (0.65*1000*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 ton^{-1} mixture input)	DM from solid fraction (kg ton^{-1} mixture input)	Total DM (kg ton^{-1} mixture input)	Total water[a] (kg ton^{-1} mixture input)	Total heat consumption (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 (kWh ton^{-1} input mixture)		Heat (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	*Mass balance: changes during biogas production*	*Mass balance: amount after biogas production*	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	*No change*	*8.0*	8.7
Phosphorus (P)	2.9	*No change*	*2.9*	3.2
Potassium (K)	2.3	*No change*	*2.3*	2.5
Carbon (C)	85.4	*-38.1* [c]	*47.3*	51.4
Copper (Cu)	0.04	*No change*	*0.04*	0.04
Zinc (Zn)	0.2	*No change*	*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-CH_4 and C-CO_2) from the biogas plus the losses from the digestion process. In this study, CH_4 losses from the digestion process are calculated as 1 % of the

produced methane. Biogenic CO_2 losses are calculated based the biogenic methane losses (Table S11), i.e. 1.42 kg CO_2 are emitted per kg of CH_4.

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

	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
Unit	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-CH_4 and C-CO_2) from the biogas plus the losses from the digestion process. In this study, CH_4 losses from the digestion process are calculated as 1 % of the produced methane. Biogenic CO_2 losses are calculated based the biogenic methane losses (Table S11), i.e. 1.67 kg CO_2 are emitted per kg of CH_4.

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	*Mass balance: amount transferred to the degassed solid fraction (deg.SF)*	*Mass balance: amount transferred to the degassed liquid fraction (deg.LF)*	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	*Mass balance: amount transferred to the degassed solid fraction (deg.SF)*	*Mass balance: amount transferred to the degassed liquid fraction (deg.LF)*	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 kg P per ton slurry * 25.48 ton slurry ha^{-1} = 26.50 kg P per ha
	K	2.60 kg K per ton slurry * 25.48 ton slurry ha^{-1} = 66.25 kg K per ha
Cow	P	0.98 kg P per ton slurry * 30.78 ton slurry ha^{-1} = 30.16 kg P per ha
	K	5.65 kg K per ton slurry * 30.78 ton slurry ha^{-1} = 173.91 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
Annual average (kg ha^{-1})		*21.5*	*64*
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
Annual average (kg ha^{-1})		*25.4*	*131.0*

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.

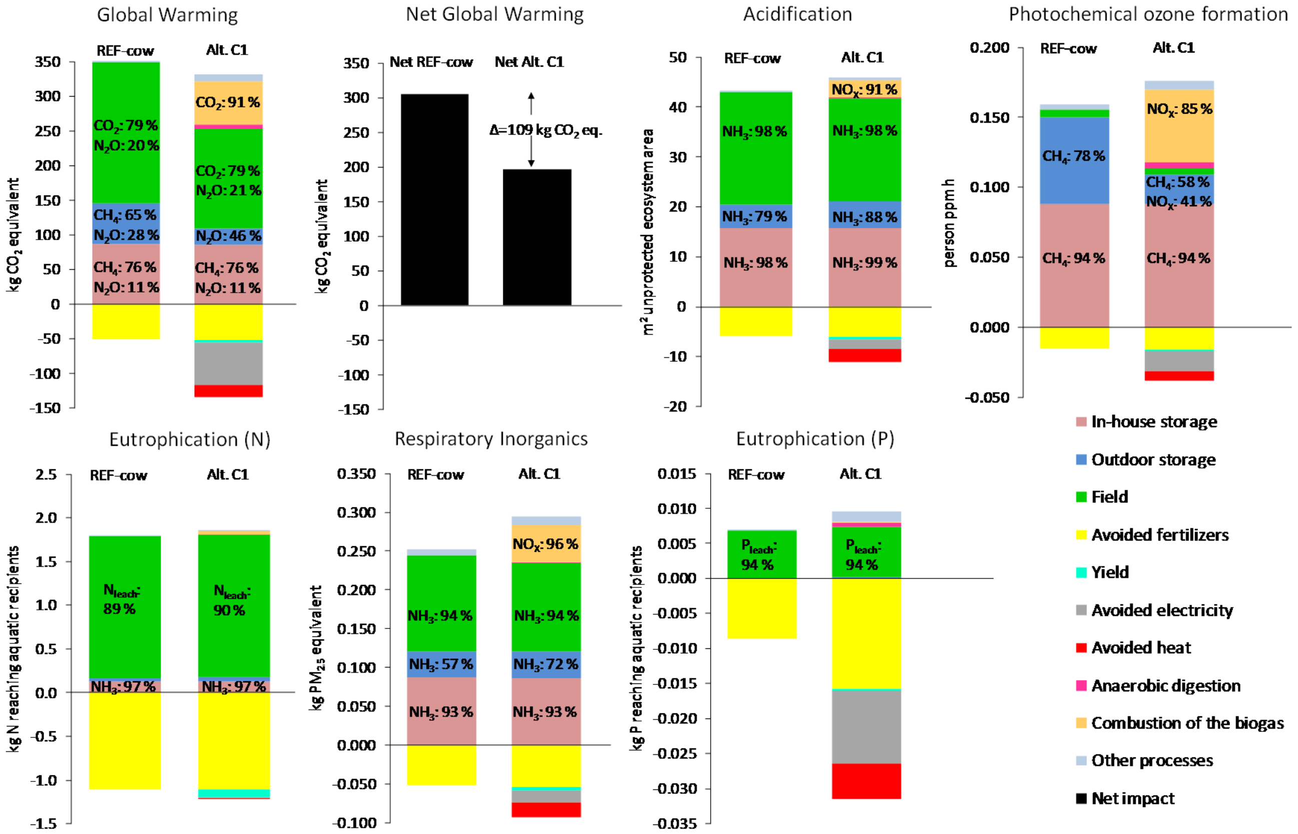

Figure S2. Breakdown of impact assessment results for all impacts, for alternative C1

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 CO_2 through the molecular weight ratios. It does not represent a sequestration of CO_2 (it is C that is sequestrated).

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 ($CH_2CHCOOH$), which in turn is degraded to CO_2 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 °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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A.2. Article 2: LCA of biomass-based energy systems: a case study for Denmark

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LCA of biomass-based energy systems: a case study for Denmark

Tonini, D. & Astrup, T.*

Department of Environmental Engineering – Technical University of Denmark
DTU-Building 115, DK-2800 Kgs. Lyngby, Denmark

*Corresponding author: Davide Tonini
E-mail: dait@env.dtu.dk

Abstract

Decrease of fossil fuel consumption in the energy sector is an important step towards more sustainable energy production. Environmental impacts related to potential future energy systems in Denmark with high shares of wind and biomass energy were evaluated using life-cycle assessment (LCA). The results showed that significant reductions of greenhouse gas emissions could be achieved by increasing the energy production from wind and biomass resources, although additional cultivation of perennial crops was required to cover the energy demand. Production of biofuels for heavy transportation represented a challenge, as cultivation of oil-plants (e.g. rapeseed) caused large environmental impacts (global warming) due to land use changes (LUC), and the thermochemical pathway (Fischer-Tropsch) showed impacts similar to consumption of fossil diesel. The energy systems based on extensive cultivation of energy crops were associated with significantly increased impacts related to aquatic eutrophication due to increased fertilizer consumption. Land occupation increased to a range of 600-2100 ha/PJ depending on the amounts and types of energy crops introduced.

Keywords: LCA, LUC, biomass potential, energy system analysis, environmental impacts

1. Introduction

In many countries, considerable efforts are being made to reduce greenhouse gas (GHG) emissions within the energy sector as part of the response to climate changes. Within the recent decades, Denmark has managed to control the energy demand which today is similar to that before the oil crisis in the 1970s (864 PJ). Today the share of oil in the system corresponds to 20% and 20% of the electricity production is based on wind. Overall, 15% of the primary energy supply is based on renewables such as biomass, solar energy and waste resources [1]. The long-term target for Denmark is to reach a 100% renewable energy system. Several studies have modeled future sustainable energy systems from a technical perspective [2-7]. According to these studies, 100% renewable energy systems can only realistically be achieved through significant reductions in energy demand, increased efficiencies of fuel conversion technologies, higher shares of wind power (e.g. up to 50%), replacement of fossil fuels with biomass resource and integration of the transport sector into the energy system e.g. through establishment of electric vehicles [8,9]. Although the primary focus of studies involving energy system analysis is on the technical design of the energy system (modeling of energy demand

and supply, fuels requirements, and technology implementation), many of these studies also report associated CO_2 emissions as an indicator for the environmental impacts related to the energy system in question. However, such calculations of direct emissions associated with the combustion of fuels do not account for important upstream or downstream environmental impacts related to the energy system, for example land use changes (LUC, due to energy crops cultivation), cascading effects (e.g. substitution of products in the market with byproducts form biofuel production), utilization of residues (e.g. digestate and biochar).

GHG emissions have received considerable attention recently; however, other potential environmental impacts are associated with energy production (e.g. eutrophication, acidification and land use). Such impacts are generally not considered by energy system analysis. To provide a full overview of the environmental consequences of changing energy production in the future, all upstream, direct and downstream emissions have to be accounted in a life-cycle perspective. We have found no such studies in the literature focusing on energy systems with high shares of wind and biomass energy.

This study quantifies the environmental impacts associated with selected future energy systems for Denmark (2008, 2030, and 2050). Life-cycle assessment (LCA) was used to quantify these impacts. All relevant energy technologies and processes in the energy system were addressed (e.g. wind energy, hydropower, photovoltaic, solar and biomass-to-energy technologies); however, particular focus was placed on the biomass-to-energy subsystems as biomass and associated land use effects were a specific concern. The specific objectives were: i) identification of potential biomass resources, ii) identification and selection of suitable biomass conversion technologies and related efficiencies, and iii) quantification of environmental impacts associated with the selected energy systems. Results were evaluated relative to the impact categories: global warming, acidification, aquatic eutrophication and land occupation.

2. Methodology

2.1 Goal, scope and functional unit

The goal of the LCA was to assess the environmental impacts related to more sustainable energy systems in Denmark. A range of potentially future energy systems were selected based on a mix of residual agricultural resources, energy crops, wind and other renewables (e.g. waves and solar energy) and to the extent needed also fossil fuels. All future energy systems were compared with a reference scenario representing the Danish energy system in 2008. The LCA functional unit was "production of energy (electricity, heat and fuels) required in the years 2008, 2030 and 2050". The energy mix (share of electricity, heat and fuel) is shown in Table 1. As the energy demand was not identical in all years, the modeling results were normalized with the primary energy demand for the individual years to allow comparison. Thus, the results were expressed as environmental impact per unit of primary energy demand (e.g. ktonne CO_2-eq/PJ). The environmental impacts were quantified for a time horizon of 100 years according to common practice [10].

Table 1

2.2 Impact assessment

The life-cycle assessment was carried out according to the EDIP 2003 method [10] for the environmental impact categories: global warming, acidification and aquatic eutrophication, whereas for the impact category land occupation, the IMPACT 2002+ method was used [11]. For multiple-output processes like biorefineries, where valuable byproducts (e.g. fodder or chemicals) were generated together with fuels, system expansion was applied and it was assumed that these products substituted the marginal products in the market, according to the principles of consequential LCA [12]. The life-cycle assessment was facilitated by the LCA-software Simapro 7.1 [13].

2.3 Energy scenarios

Five different energy systems were assessed: I) 2008 (reference), II) 2030, III) 2050CSV, IV) 2050RME and V) 2050BtL. The latter three represented different alternatives for transport fuel production in year 2050. The “2008” system was selected as reference representing the current energy system primarily based on fossil resources. Data for energy demand and supply were based on Danish national statistics for 2008 [14]: the gross primary energy demand was 864 PJ while the final net consumption by society was 660 PJ. An overview of the energy system “2008” is shown in Figure 1.

Energy systems representing 2030 and 2050 were associated with significant reductions in energy demand and based on improved efficiencies of CHP plants, increased electricity production from wind energy, replacement of fossil fuels with biomass and the introduction of electric vehicles. The technical aspects of these systems have been discussed elsewhere [3].

The “2030” system represented a link between 2008 and the 2050 systems: about 50% of the energy was generated from renewable resources. The transition to electric vehicles was assumed to be incomplete and therefore ethanol was required as fuel in the energy system [3]. In order to fulfill the demand for heat and electricity, about 40 PJ of energy crops (willow) were assumed cultivated in addition to the estimated biomass potential resources. The gross primary energy demand was estimated to 710 PJ while the final net consumption by society was 534 PJ. The higher efficiency of the energy system was mainly due to technical measures assumed to be implemented for reduction of the total energy demand [3]: e.g. decommissioning of old inefficient power plants, construction of new more efficient power units (utilizing fuel cells), improvement and expansion of district heating networks, and insulation of buildings. An overview of the energy system “2030” is shown in Figure 2.

For 2050, three different alternatives for production of fuels in the transportation sector were assessed (all other aspects of the three energy systems (industry, power plants, household) were identical, see Figure 3). In order to fulfill the energy demand, about 51 PJ of willow were required to be cultivated in addition to the estimated biomass potential resources. Most of the transportation was based on electricity produced from renewables (also identical in the three systems), except for heavy vehicles and aviation which required diesel and long-chain hydrocarbons (kerosene and aviation fuel). The final net energy consumption by society was 459 PJ. Technical measures, similar to those in 2030, for reduction of the overall energy demand were included. In “2050CSV”, 63 PJ of crude oil was assumed to fulfill the demand for heavy terrestrial transportation (about 35 PJ) and aviation (about 33 PJ). The gross primary energy demand was 552 PJ. In “2050RME”, 35 PJ rape methyl ester (RME) was assumed to fulfill the fuel demand of heavy terrestrial transportation supplemented by 33 PJ of crude oil for aviation. The gross primary energy demand was 576 PJ. In “2050BtL”, 30.5 PJ of Fischer-Tropsch (FT) based biodiesel from willow and 4.5 PJ of RME was assumed for terrestrial transportation supplemented by 33 PJ of crude oil for aviation. The gross energy demand was 587 PJ.

Figure 1

Figure 2

Figure 3

2.4 Life cycle inventory data

Biomass resources

The relevant biomass resources available in Denmark were: manure, grass, lignocellulosic biomass (e.g. wood and straw) and waste. The total amount of biomass potential was estimated to be about 182.3 PJ. Focus is on residual biomass, i.e. waste (e.g. municipal solid waste, MSW) and byproduct/residues from agriculture and forestry (e.g. straw, manure, wood). Today most biomass resources have a function in the ecosystem or in the economy meaning that the utilization of these resources for energy production would induce changes in the ecosystem or in the society if status quo is to be maintained. As a consequence, the use of biomass resources for energy purposes instead of the current use (e.g. feeding, bedding, ploughing back to fields etc.) will finally lead to a competition between energy and other uses. The consequences of routing biomass resources to energy production were addressed in the LCA. A detailed description of the biomass potential for Denmark is reported in SI.1.

Energy conversion technologies

The choice of the biomass-to-energy conversion technologies was based on a number of considerations which implied energy system as well as technical issues for handling the biomass (see SI.2). The production of an intermediate energy carrier (e.g. biogas and syngas) was preferred to direct combustion for the flexibity and storability of the energy products which are needed to accommodate the fluctuations of energy systems with high penetration of wind power [15]. Manure and grass were assumed to be fermented to biogas through anaerobic digestion processes. Lignocellulosic biomass (e.g. wood, straw and willow) was assumed to be gasified for syngas generation. Biogas and syngas were then converted to heat and electricity in solid oxide fuel cells (SOFC) with high electricity efficiency. Waste was assumed to be incinerated for heat and electricity production. Biodiesel was produced from rapeseed and willow by means of transesterification and thermal process (gasification and Fischer-Tropsch), respectively.

Table 2 provides an overview of the background life-cycle inventory (LCI) data for (selected) biomass-to-energy (and to-fuel) processes used in the assessment. With respect to the LCIs for wind, hydro and wave power, heat pumps, SOFC, fossil fuel combustion in combined heat and power (CHP) plants, district heating plants, peak-load boilers, vehicles, offshore platforms and industrial furnaces for heat production, common processes found in the Ecoinvent database [16] were used. A detailed description of the energy conversion technologies is reported in SI.2.

Table 1

Land use changes

Cultivation of energy crops requires use of land thereby inducing direct and indirect land use changes (dLUC and iLUC) under the basic assumption that land available for cultivation is constrained.

With respect to willow, the dLUC were estimated based on [17]. The iLUC were estimated based on the assumption that expansion of willow cultivated land in Denmark replaced the marginal crop (spring barley) which had to be produced somewhere else if status quo was to be maintained. The most likely consequence was assumed to be conversion of grassland into barley (69%) as well as intensification of barley cultivation in Canada (31%) [40]. The land use consequence of replacing prairie grass with barley was 84 tonne CO_2/ha. Intensification implied a larger utilization of fertilizers in order to increase the production on the same constrained land (1 kg N/ha for Canadian conditions).

With respect to rapeseed, dLUC and iLUC were quantified according to [18] assuming conversion of set-aside land into rapeseed (all 2050 scenarios) or conversion of set-aside land and arable land (spring barley) into rapeseed (only the “2050RME” scenario). For conversion of arable land (spring barley) into rapeseed, a carbon loss of 0.115 tonne C/ha/y was assumed according to [17]. Only dLUC and iLUC associated with changes in rapeseed cultivation from the current situation to the future needs were considered.

The uncertainties in the assumptions were addressed in the sensitivity analysis. Table 3 provides an overview of the background data used to evaluate dLUC. A detailed discussion of the impacts associated with land use changes is reported in SI.3.

Table 2

Management of agricultural and biomass conversion residuals

The removal of straw from fields induces changes in the soil carbon stock. The calculated carbon depletion was 0.09 tonne C/tonne straw [19]. Removal of nutrients (N, P and K) with the straw led to additional fertilizer use to maintain constant crops yields. Straw removal also caused lower N_2O emissions: a decrease of 0.03 kgN-N_2O/tonne DM straw was assumed [19].

The use of grass for energy instead of feeding induced an increased demand for other types of fodder. This was modeled with additional production of barley in order to satisfy the feed demand.

The use-on-land of digestate from anaerobic digestion of manure was credited by substitution of inorganic N, P, K fertilizers [20]. Application of 1 tonne of digestate was assumed to substitute 4.07 kg of ammonium nitrate (as N), 2.1 kg of triple superphosphate (as P_2O_5) and 3.3 kg of potassium chloride (as K_2O).

The use-on-land of biochar was also credited for its potential positive effects on soil, e.g. carbon sequestration, improved fertilizer efficiency and reduced N_2O emissions, based on [21]. A detailed description of the assumptions regarding management of agricultural and biomass conversion residuals is reported in SI.4.

3. Results and discussion

The results are presented with respect to the environmental categories global warming (GW), acidification (AC), aquatic eutrophication (AE) and land occupation (LO) in Figure 4. The impacts were calculated per unit of energy provided to society (e.g. ktonne CO_2-eq/PJ). The results for the category land occupation are presented as additional land required (Δha/PJ) compared with the current situation (“2008”). Only the sub-processes (e.g. transportation, LUC, fossil fuel combustion etc.) contributing to the overall impacts with more than 1% are shown. Figure 5 shows the environmental impacts associated with the production and combustion of RME and FT-biodiesel compared with traditional diesel. In this case the results were re-calculated corresponding to a functional unit of 1 energy unit of diesel-fuel for the purpose of comparison. Lastly, Table 4 shows the numerical results for normalized and total environmental impacts.

3.1 Global warming (GW)

Overall the results for GW indicated a decreasing trend for GHGs emissions from 2008 (about 90 ktonne CO_2-eq/PJ) to 2050 (about 18-31 ktonne CO_2-eq/PJ depending on the scenario). The reduction of GHGs emissions (per PJ primary energy) was thus in the range of 66%-80%. The preferred scenarios from a GW perspective were the “2050BtL” and “2050CSV” scenarios whereas the worst was the “2050RME” scenario. The difference among these scenarios was caused by the magnitude of iLUC's. The impacts associated with rapeseed cultivation were significantly higher than those for willow due to the low yield and hence higher iLUC. This demonstrated that significant iLUC's associated with energy crops cultivation can completely off-set the benefits of biofuels. In this context the use of diesel for heavy transport was still favorable over RME, whereas FT-biodiesel production (the “2050BtL” scenario) showed slightly lower GW impacts than fossil diesel. The latter result, however, strongly depended on the assumptions (biochar effects and willow yield). The impacts associated with dLUC/iLUC were estimated to respectively 5, 18 and 8 ktonne CO_2-eq/PJ in the “2050CSV”, “2050RME” and “2050BtL” scenarios. It has to be noted that use-on-land of digestate and biochar led to significant GW savings owing to the return of nutrients and carbon to the soil.

3.2 Acidification (AC), aquatic eutrophication (AE) and land occupation (LO)

The results for AC followed the trends observed for GW. Decreased NO_x and SO_x emissions from fossil fuel combustion in power plants lowered the impacts compared to “2008”. The optimal scenario was “2050CSV” contributing with a load of 122 ha/PJ, while the “2050BtL” scenario at 135 ha/PJ was second best. The environmental load was mainly associated with tailpipe emissions of NO_x from biodiesel combustion in heavy vehicles (corresponding to about 58 ha/PJ). Biodiesel-fuelled heavy vehicles generally have higher NO_x emissions than conventional diesel-fuelled vehicles [22-25], which also was the reason for the high AC impacts in the “2050RME” scenario. Among the 2050 scenarios, the worst environmental performance was achieved by “2050RME” (178 ha/PJ) where cultivation of rapeseed contributed with 34 ha/PJ (N-fertilizers) in addition to the NO_x related impacts.

All the assessed scenarios contributed with significant impacts in the AE category, mainly associated with the increased use of fertilizers for energy crop production and the consequent release of nitrates and phosphates to surface waters. The least preferable scenario was “2050RME” (29 tonne N/PJ, double of the “2008” scenario) due to the high amount of fertilizers required for rapeseed and barley cultivation whereas cultivation of willow in “2050BtL” required less fertilizers. This was in agreement with several other studies, e.g. [26]. The AE impacts associated with transportation was higher in the scenarios including biodiesel-fuelled heavy vehicles because of higher NO_x tailpipe emissions, in accordance with the results for AC.

The “2050RME” and “2050BtL” scenarios required the largest area of land (respectively additional 2092 and 1790 ha/PJ compared with “2008”). This was caused by cascading effects due to the cultivation of energy crops in Denmark and subsequent displacement-replacement mechanisms as previously mentioned. The scenarios “2030” and “2050CSV” required significantly less additional land due to use of fossil fuels for heavy terrestrial transportation in place of biodiesel.

Figure 4

3.3 Impact of biodiesel production

RME-biodiesel was by far the least desirable option with respect to all the environmental categories. With respect to GW, the impact was estimated to 287 g CO_2-eq/MJ of fuel (i.e. ktonne CO_2-eq/PJ), whereas for fossil diesel the corresponding value was about 89 g CO_2-eq/MJ. These results are in agreement with other findings in literature (e.g. [27,28]). The impacts from FT-biodiesel were in the range of 65-88 g CO_2-eq/MJ depending on assumptions regarding biochar (see sensitivity analysis). RME-biodiesel was also the least favorable option in relation to the AC, AE and LO categories. With respect to AC and AE, the loads were mainly associated with tailpipe emission of NO_x and use of N-fertilizers for crop cultivation as previously explained. This was also the case for FT-biodiesel. However, the impacts associated with cultivation of willow for FT-biodiesel production was significantly less due to the higher yield and reduced fertilizer use.

Figure 5

3.4 Sensitivity analysis

The sensitivity of the results towards changes in assumptions and parameters was carried out in order to assess the significance of: I) willow yield, II) magnitude of dLUC/iLUC associated with cultivation of rapeseed, III) efficiency of BtL processes and IV) biochar effects on GW. These aspects were identified as having the highest potential for affecting the overall conclusions. I) The yield of willow was varied between 7 and 16 tonne DM/ha which is a likely range for Denmark [29]. II) The iLUC associated with rapeseed were estimated according to [27]. An average value of 1.32 kg CO_2/kg rapeseed was assumed (only effects on GW was assessed as no information on impacts in other impact categories were available). III) The efficiency of the BtL process was set to 57% with use of hydrogen generated from electrolysis of wind power (excess wind power was assumed to be available). IV) No benefits for GW from biochar were considered. Results from the sensitivity analysis are presented in Table 5.

A low yield for willow (I) made the use of fossil fuel for heavy transport favorable to FT-biodiesel produced from lignocellulosic biomass, according to all the investigated impact categories. A new value for iLUC (II) changed the overall result for GW for the “2050RME” scenario. The impacts associated with iLUC significantly decreased (Δ=-11 ktonne CO_2-eq/PJ) compared with the baseline scenario (based on [18]). However, the different approach for estimation of iLUC did not affect the overall ranking of the 2050 scenarios: the scenario based on RME biodiesel was still the least favorable.

More efficient thermochemical processes (III) combined with electricity supply from wind power only slightly improved the environmental performance of biofuel production via thermochemical conversion, except with respect to LO. Although, several technologies for utilization of excess wind power in future energy systems exist, it should be noted that constraints (e.g. capacity and interconnectors) in the electricity system may be limiting. If the benefits of biochar (IV) were not included, the performance of the “2050BtL” scenario became similar to the “2050CSV” scenario, i.e. FT-biodiesel did not contribute with saving in the GW category compared with fossil diesel.

Overall, the sensitivity analysis revealed that quantification of LUC impacts, assumption regarding yields of energy crops and potential benefits from biochar use can significantly affect the overall result of the LCA. However, despite these effects, the overall ranking of the individual scenarios did not change. The overall results based on the assessed scenarios are therefore considered robust.

Table 5

4. Conclusion

The environmental impacts related to four potentially future energy scenarios for Denmark were compared with the energy system in 2008 by means of LCA. It was demonstrated that: i) residual domestic biomass resources were insufficient to cover the energy demand thereby requiring cultivation of energy crops, ii) high impacts associated with LUC made the use of fossil diesel for heavy transport favorable to RME biodiesel, iii) high potential nutrient enrichment effects were a direct consequence of energy crops cultivation practices. The results showed that significant reductions in GHG emissions can be obtained by increasing the share of wind power and utilizing lignocellulosic biomasses (willow, wood and straw by gasification and grass and manure by anaerobic digestion with subsequent conversion of the gas to electricity and heat). However, by far the main "challenge" was provision of biofuels for heavy terrestrial transport and aviation. Aviation still relied on fossil fuel since no mature technology for production of aviation biofuel was found in literature. With respect to terrestrial transportation, low efficiencies of transesterification and thermochemical processes led to significant environmental impacts related to global warming (mainly LUC), aquatic eutrophication (increased fertilizers use) and land occupation. The use of RME biodiesel for heavy terrestrial transport was the least favorable option with respect to the environmental impacts.

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Tables

Table 2 Energy demand in the reference years (rounded values)

Energy demand (PJ)	2008	2030	2050CSV	2050RME	2050BtL
Primary energy	864	710	552	576	587
Electricity	187	97	94	94	94
Heat	239	294	284	284	284
Fuel for transport	220	144	80	80	80
’Non-energy’ use	14	-	-	-	-
Energy consumed by society	660	534	459	459	459

Table 3 Overview of the background data used in the LCI of energy conversion technologies for selected biomass-to-energy (or -fuel) processes. BtL: Biomass-to-Liquid process; EtOH: (bio) ethanol

Biomass	Energy technology	Products	Use of products	LCI (source)
Manure	Anaerobic digestion	Biogas & digestate	Biogas to heat & electricity. Digestate to use on-land	[16,20]
Grass	Anaerobic digestion	Biogas, solid biofuel & proteins	Biogas to heat & electricity. Grass fibers to heat & electricity. Proteins substitute soymeal	[16]
Wood & willow	Gasification	Syngas & biochar	Syngas to heat & electricity. Biochar to use on-land	[16,30-32]
Straw	Gasification	Syngas & biochar	Syngas to heat & electricity. Biochar to use on-land	[16,33]
Waste	Incineration	Electricity & heat	-	[16,34]
Rapeseed	Transesterification	RME, glycerin & solid biofuel	RME to transport. Glycerin substitutes glycerin production. Biofuel to heat & electricity	[16]
Willow (BtL)	Gasification and Fischer & Tropsch	FT-diesel & biochar	FT-diesel for transport. Biochar to use on-land	[35]
Straw (EtOH)	Straw refinery	EtOH, molasses & solid biofuel	Bioethanol for transport. Molasses substitutes fodder. Biofuel to heat & electricity.	[16,36]

Table 4 Overview of the background data used in the LCI for the effects associated with direct land use changes (dLUC) for selected crops. Negative values indicate emissions (e.g. loss of carbon), positive values indicate sequestration

Crop	tonne CO_2/ha	tonne N_2O/ha	tonne NO_3/ha	LCI data (source)
Barley	-84	-0.02	-4.6	[18]
Rapeseed	-88	-0.022	-4.6	[18]
Willow	0.12	-0.0026	-2.3	[17,20,37]

Table 5 Environmental impacts (normalized and total results) for the selected energy systems

			Unit	Energy system				
				2008	2030	2050CSV	2050RME	2050BtL
Energy mix	Primary energy		PJ	864	710	552	576	587
	Electricity			187	97	94	94	94
	Heat			239	294	284	284	284
	Fuel (transport)			220	144	80	80	80
Impacts	GW	Normalized	ktonne CO_2-eq/PJ	68	36	20	31	18
		Total	Mtonne CO_2	59	26	11	18	10
	AC	Normalized	ha/PJ	301	184	122	178	135

	Total	10^3ha	260	130	68	100	79
AE	Normalized	tonne N/PJ	14	15	15	29	24
	Total	ktonne N	12	11	8.4	17	14
LO	Normalized	Δha/PJ	-	591	872	2092	1790
	Total	10^3Δha	-	420	480	1200	1100

Table 5 Sensitivity analysis: results are expressed as net difference ‘Δ’ compared to the baseline result (e.g. Δktonne CO_2-eq/PJ, Δha/PJ, Δtonne N/PJ, etc.). The sensitivity analysis on RME (II) and biochar (IV) only affected GW category (y: yield; η: efficiency; ↑: increase; ↓ decrease)

Category	Parameter	2050CSV	2050RME	2050BtL
GW	(I) Willow (y↑ / y↓)	-1/+2	-1/+2	-2/+4
	(II) RME (iLUC ↓)	-	-11	-
	(III) BtL (η ↑)	-	-	-1
	(IV) Biochar (↓)	+2	+2	+4
AC	(I) Willow (y↑ / y↓)	-1.6/+4	-1.6/+4	-5/+7
	(III) BtL (η ↑)	-	-	-2
AE	(I) Willow (y↑ / y↓)	-1/+2.5	-1/+2.5	-3/+4.5
	(III) BtL (η ↑)	-	-	-1
LO	(I) Willow (y↑ / y↓)	-60/+443	-60/+443	-445/+633
	(III) BtL (η ↑)	-	-	-101

Supporting Information (SI) for:

“LCA of biomass-based energy systems: a case study for Denmark”

Tonini, D.* & Astrup, T.

Technical University of Denmark, Department of Environmental Engineering

* Corresponding author email: dait@env.dtu.dk

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

SI.1 Biomass resources
SI.2 Energy conversion technologies
SI.3 Land use changes
SI.4 Management of agricultural and biomass conversion residuals

SI.1 Biomass resources

Relevant biomasses resources available in Denmark (Table S1) were: manure, grass, lignocellulosic biomass, waste, beet top, molasses, whey, potato pulp, brewer's grain. With respect to the straw, today only 41% and 15% of straw from corn, barley and rape (not used for feeding or bedding) is used for energy production. The remaining is ploughed back to the field. According to [1], if 100% removal and utilization was assumed the total potential of corn, barley and rape derived straw would be about 39 PJ. In this study a straw potential of 39 PJ was assumed. The environmental consequences of straw removal were included in the LCA.

The estimates behind the data reported for energy crops (9.1 PJ) were based on the assumption that 50% of the land currently "lying fallow" can be used for energy crops, such as willow or mischantus [1]. The remaining 50% of land was considered unsuitable for crop cultivation as the soil quality was poor. These low-lying areas were instead assumed suitable for cultivation of grass. All of this grass (100% exploitation) was assumed utilized for biogas production yielding about 6.8 PJ of biogas. The overall energy potential of grass was estimated equal to about 13 PJ based on a HHV of 18 GJ/tonne FM and on a biogas yield of 400 Nm^3/tonne DM. This was done in order to include the energy potential of the fibers residue (i.e. solid biofuel to be combusted) left after anaerobic digestion. The consequences of less grass available for feeding were included in the LCA.

Waste quantities were predicted to increase to 47 PJ in 2030, following the trends from previous years. From 2030 to 2050 no increase was included, assuming that the share of recycling increases thereby leaving less waste available for energy purposes. Waste quantities only included waste which is today used for energy purposes. In this study, beet top, molasses, whey, potato pulp and brewer's grain were disregarded because these resources are today for animal feeding and/or bedding and because of the relatively small energy potential associated with the resources.

The total quantities of biomass potential resources (182.3 PJ) did not match the energy demand in the future scenarios, therefore cultivation of energy crops had to be considered. About additional 51 PJ of willow were previously estimated to be required to fulfill the needed electricity and heat demand [2]. Willow was selected as a favorable lignocellulosic energy crop among the others (e.g. mischantus, poplar etc.) because of the high yield, low requirement of fertilizers and other agricultural practices, capacity of sequestering carbon, adaptability to different soils etc. [3,4]. However, the choice of mischantus or other short rotation coppice (SRC) would not affect the results, as yield and fertilizers needs for these crops are similar [4]. Rapeseed was selected as oil crop for production of biodiesel, as rapeseed cultivation is already practiced in Denmark and because of lower environmental impacts compared with the cultivation of soy.

Table S1 Overview of the biomass potential for Denmark. The values are expressed as primary energy (LHV) before energy conversion, except of manure and grass (in brackets) for which potentials are expressed as energy in the biogas. U: in use; P: potential

Biomass (PJ)	U [5]	P [5]	U [6]	P [6]	P [1]	P(this study)
rapeseed	3.4	4.5	-	-	4.5	4.5
willow	0.5	9.1	-	-	9.1	9.1
grass	0	5.1	-	-	5.1	13 (6.8)*
straw	18.5	26.8	17.3	26.8	33.5	39
beet top	-	-	-	-	0.2	-
animal manure	1.1	20.2	-	-	20.2	27
fiber fraction	0	2.5	-	-	2.5	2
mill residues	-	-	-	-	0.9	-
beet pulp	-	-	-	-	1.7	-
molasses	-	-	-	-	1.2	-
potato pulp	-	-	-	-	0.3	-
brewer's grain	-	-	-	-	0.6	-
whey	-	-	-	-	2.8	-
wood chips	-	-	9.8		7.7	9.8
fire wood	-	-	23		26	23
unexploited forest increment	-	-	-	40	17	-
wood pellets	-	-	2.3		2.6	2.3
wood residues	-	-	5.6		6.3	5.6
waste	-	-	23	34-41	-	47
paper and cardboard	-	-	5-6	5-6	-	-
industrial waste	0.9	1.5	-	-	-	-
animal fat	1.9	3.2	-	-	-	-
meat and bones	0	1.6	-	-	-	-
Total						182.3

* The overall energy potential of grass was estimated equal to 13 PJ based on a HHV of 18 GJ/tonne DM (see Figure 2-3). This was done in order to include the energy potential of the fibers residue left after anaerobic digestion.

SI.2 Energy conversion technologies

The choice of the biomass-to-energy conversion technologies was based on the following considerations which implied energy system as well as technical issues for handling the biomasses.

Future energy systems in Denmark will have to face the challenge of integrating high shares of fluctuating energy sources, such as wind power, into the energy system. In order to reach this goal more flexible biomass conversion technologies are needed to accommodate the fluctuations [7]. Production of intermediate energy carriers (e.g. syngas or biogas) may therefore be favorable to direct combustion of biomass, at least in those cases where the overall conversion efficiency is competitive with direct combustion. Anaerobic digestion for biogas production is already widely applied in Denmark [8]. With respect to lignocellulosic biomass, thermal gasification is a promising technology for conversion to gas with as high as 90% cold gas efficiencies (CGE), e.g. [9-11]. As minimization of transport distances is important, installation of local small-scale facilities for conversion of biomass should be preferred over centralized plants (provided efficiencies are competitive). Thermal gasifiers have been demonstrated to be competitive with combustion plants at a small-scale (100-600 kWe) [9-13]. Lignocellulosic biomass was therefore assumed gasified for production of syngas with efficiencies as reported in Table S3. Production of biochar was estimated to 20 kg/tonne lignocellulosic biomass [14]. Biochar was assumed to be returned to soil and the energy system was credited for the carbon sequestration and other benefits.

Manure as well as grass was assumed to be fermented to biogas through anaerobic digestion with efficiencies as reported in Table S2. Manure was assumed to be separated into a liquid and solid fraction through centrifugal separation combined with addition of cationic polymer [15-18]. The solid fraction was then added to raw manure to boost biogas production. Overall, such process increased N and P availability in the residual digestate after anaerobic digestion [15,16]. With respect to grass, the multiple-output grass-refinery process [19] generated biogas (500 Nm^3/tonne DM grass), grass fibers (0.4 tonne/tonne DM grass) and proteins (0.15 tonne/tonne DM grass). The grass fibers were assumed to be combusted in cogeneration plants producing electricity and heat [20]. The proteins were assumed to substitute for soy meal. The substitution ration was: 1 kg of proteins substituted for 1 kg of soy meal based on the proteins content. Syngas and biogas were assumed used in Solid Oxide Fuel Cells (SOFC) after upgrading to natural gas quality. Solid waste was assumed combusted in waste incinerators with electricity and heat efficiencies of 25% and 70%, respectively (typical values for Danish incinerators) [21].

Rapeseed was assumed converted into Rape Methyl Ester (RME) through a transesterification process in modern biorefineries [19]. The relevant process outputs from 1 tonne of rapeseed were: RME (0.35 tonne), rape meal (0.6 tonne) and glycerin (0.038 tonne). 1 kg rape meal was assumed to substitute soybean (0.76 kg) and spring barley (0.11 kg) [22]. 1 kg glycerin was assumed to substitute glycerin produced from fossil resources on a 1:1 ratio. The “2030” scenario involved production of bioethanol from straw [23,24] with the following outputs per tonne DM straw: ethanol (0.21 tonne), C5 molasses (0.254 tonne, 30% water content) and solid biofuel (0.35 tonne, 10% water content). Molasses were assumed to substitute spring barley (0.96 kg) [24].

Production of biodiesel through the Biomass-to-Liquid pathway (BtL) was assumed to be based on Fischer-Tropsch technology and processes [25]. Overall Biomass-to-Liquid conversion efficiencies depended on process configurations and biomass types but varied between 35% (electricity is co-generated) and 57% (maximum fuel production) for lignocellulosic biomasses. A Biomass-to-Liquid conversion efficiency of 45% was assumed. The processes required for FT-biodiesel production were: gasification of biomass, gas cleaning, gas conditioning and compression, Fischer-Tropsch synthesis and final refinery. Electricity consumption was 0.035 kWh/MJ FT-diesel (gasification), 0.066 kWh/MJ FT-diesel (gas cleaning), 0.013 kWh/MJ FT-diesel (gas conditioning and compression), 5E-06 kWh/MJ FT-diesel (Fischer-Tropsch). For the final refinery, common processes for fossil fuels refining found in the Ecoinvent database [25] were used. The electricity consumption in modern refineries is about 0.001 kWh/MJ products. The production of FT-diesel through thermochemical conversion is thus a very energy intensive process and the extra electricity and heat required in the system compared with traditional refinery of fossil fuel was accounted for as diminished efficiency (i.e. as extra consumption of biomass in the process). The overall efficiency of the BtL process (accounting for the extra biomass) was about 40%.

[16,19]With respect to the LCIs for wind, hydro and wave power, heat pumps, SOFC, fossil fuel combustion in combined heat and power (CHP) plants, district heating plants, peak-load boilers, vehicles, off-shore platforms and industrial furnaces for heat production, common processes found in the Ecoinvent database were used.

Table S2 Biogas (or CH_4) potential of anaerobic digestion technologies for grass and manure (FM=fresh matter; DM=dry matter; VS=volatile solids)

Biomass	Unit	Value	Source	Note	This study
Manure	Nm^3/tonne FM	27.7	[15,16]	LCA slurry management	28 Nm^3/tonne FM
	Nm^3/tonne FM	22	[26]	Data from Danish plants	
	Nm^3/tonne FM	19.8	[19]	Data from Swiss plants	
Grass	Nm^3/tonne DM	299-1080	[27]	Review	400 Nm^3/tonne DM
	Nm^3/tonne DM	210	[19]	Grass biorefinery	
	Nm^3/tonne FM	21	[28]	Estimation	
	Nm^3/tonne FM	211	[29]	Pilot-scale fermentation	
	Nm^3CH_4/tonne VS	230-350	[30]	LCA/energy analysis	
	Nm^3/tonne VS	600	[31]	Lab-test (mesophilic)	
Willow	Nm^3/tonne VS	360	[32]	Pretreatment (wet oxidation)	-
	Nm^3/tonne VS	200	[32]	Without pretreatment	
Mischantus	Nm^3/tonne VS	360	[32]	Pretreatment (wet oxidation)	-
	Nm^3/tonne VS	200	[32]	Without pretreatment	

Table S3 Cold Gas Efficiency (CGE) of thermal gasification technologies from different literature studies

Biomass	CGE	Source	Note	This study
Woodchips	0.93	[10][10,10]	Pilot-scale 2-stage fixed bed	0.93
	0.74-0.92	[14]	Pilot-scale fluidised bed	
	0.8	[33]	Pilot-scale fluidised bed	
	0.648	[34]	Lab-scale fixed bed	
	0.714	[35]	Lab-scale circulating fluidised bed	
	0.8-0.92	[36]	Review	
Wood pellets	0.96	[36]	Review	0.93
Wood waste	0.82	[37]	Review	0.85
	0.49-0.66	[33]	Pilot-scale fluidised bed	
Wood sawdust	0.605	[38]	Pilot-scale circulating fluidised bed	-
	0.569	[39]	Lab-scale fluidised bed	
	0.57	[36]	Review	
Straw	0.85	[40]	Pilot-scale 2-stage fixed bed	0.85
	0.85	[37]	Review	
	0.81	[14]	Pilot-scale fluidised bed	
Mischantus	0.4	[41]	Lab-scale circulating fluidised bed	-
	0.3-0.53	[42]	Lab-scale fixed bed	
	0.85	[37]	Review	
Willow	0.25-0.43	[42]	Lab-scale fixed bed	0.9
	0.85	[37]	Review	

SI.3 Land use changes

Cultivation of energy crops required use of land thereby inducing direct and indirect land use changes (dLUC and iLUC) under the basic assumption that land available for cultivation is constrained. A brief review of dLUC impacts is reported in Table S4. The iLUC are discussed in the following.

With respect to willow, the direct land use changes were estimated based on [43] which estimated the soil organic carbon (SOC) changes related to the conversion of different types of land into SRC. No carbon losses were estimated for conversion of grassland (or set-aside land) into SRC. Instead, increases in carbon stock were estimated when converting arable land into SRC (0-115 kg C/ha depending on the type of tillage for wheat. i.e. reduced or conventional). The indirect land use changes were estimated based on the assumption that expansion of willow cultivated land in Denmark replaced the marginal crop (spring barley) which had to be produced somewhere else if status quo was to be maintained [44,45]. The most likely consequence was assumed to be conversion of grassland into barley (69%) as well as intensification of barley cultivation in Canada (31%) [40]. The land use consequence of replacing prairie grass with barley was 84 tonne CO_2/ha. Given the assumed yield of willow (11.8 tonne DM/ha) and barley in Denmark and Canada (respectively, 5.2 tonne and 2.8 tonne DM/ha), this corresponded to an iLUC emission of about 1.5 kg CO_2/kg barley cultivated in Canada on converted land. Instead, intensification finally implied a larger utilization of fertilizers in order to increase the production on the same constrained land. According to [46] this led to an increase if N-fertilizer use of about 1 kg N/ha (for Canadian conditions). According to [1], the potential for energy crop cultivation in Denmark corresponded to 9.1 PJ. The current production (2009) was 0.5 PJ. It was therefore assumed that 8.6 PJ of willow were cultivated on Danish set-aside land implying negligible SOC (hence negligible dLUC) whereas the remaining amount required to satisfy the energy demand was instead cultivated at the expenses of the marginal crop (spring barley) implying both dLUC in Denmark and iLUC in Canada, as explained previously.

With respect to rapeseed, direct and indirect land use changes were quantified according to [47] assuming conversion of set-aside land into rapeseed (all 2050 scenarios) or conversion of set-aside land and arable land (spring barley) into rapeseed (only the “2050RME” scenario). For the conversion of set-aside land to rapeseed in Denmark an emission of 88 tonne CO_2/ha (4.4 tonne CO_2/ha/year), 0.022 tonne N_2O/ha (0.001 tonne N_2O/ha/year) and 4.6 tonne NO_3/ha (0.23 tonne NO_3/ha/year) was assumed.

For conversion of arable land (spring barley) to rapeseed, a carbon loss of 0.115 tonne C/ha/y was assumed according to [43]. Only dLUC and iLUC associated with changes in rapeseed cultivation from the current situation to the future needs were considered. The methodology as well as the final estimations of iLUC was characterized by significant uncertainty. This uncertainty has been addressed in the sensitivity analysis.

Table S4 Effects of direct land use changes (dLUC) on the soil organic carbon from different literature studies (SRC: short rotation coppice; OSR: oilseed rape). Negative values indicate emissions (e.g. loss of carbon), positive values indicate sequestration

Crop	Unit	Value	Note	Source	This study
Barley	tonne CO_2/ha	-84	Prairie grass to cropland	[47]	[47]
	tonne N_2O/ha	-0.02	Prairie grass to cropland	[47]	
	tonne NO_3/ha	-4.6	Prairie grass to cropland	[47]	
Rapeseed	tonne CO_2/ha	-88	Set-aside to cropland DK	[47]	[47]
	tonne N_2O/ha	-0.022	Set-aside to cropland DK	[47]	
	tonne NO_3/ha	-4.6	Set-aside to cropland DK	[47]	
	tonne C/ha/y	-0.634	Broadleaved forest to OSR	[43]	
	tonne C/ha/y	-0.115	Winter wheat to oilseed rape	[43]	
	tonne C/ha/y	-0.634	Grassland to oilseed rape	[43]	
	tonne C/ha	-9	Set-aside to cropland	[48]	
	tonne C/ha	-9	Temperate grassland to cropland	[48]	
	tonne C/ha	-13	Temperate forest to cropland	[48]	
	tonne C/ha	na	Tropical grassland to cropland	[48]	
	tonne C/ha	na	Tropical moist rain to cropland	[48]	
	tonne C/ha/y	-0.24	Straw ploughed back to soil	[49]	
	tonne C/ha/y	-0.4	Straw not ploughed back to soil	[49]	
Mischantus	tonne C/ha/y	0.62	dLUC (UK) for rapeseed and SRC	[49]	-
	tonne C/ha/y	0.115	winter wheat to mischantus	[43]	
	tonne C/ha/y	0	Grassland/broadleaved forest to SRC	[43]	
Willow (SRC)	tonne C/ha/y	0.14	dLUC (UK) for rapeseed and SRC	[49]	[43]
	tonne C/ha/y	0.12	Winter wheat to SRC	[43]	
	tonne C/ha/y	0.00	Grassland/broadleaved forest to SRC	[43]	
Salix	tonne C/ha/y	0.34	Fertilized	[50]	-
	tonne C/ha/y	0.22	Not fertilized	[50]	
	tonne N-N_2O/ha/y	1.15	Fertilized	[50]	
	tonne N-N_2O/ha/y	0.57	Not fertilized	[50]	
Populus	tonne C/ha/y	0.53	Fertilized	[50]	-
	tonne C/ha/y	0.23	Not fertilized	[50]	
	tonne N-N_2O/ha/y	1.99	Fertilized	[50]	
	tonne N-N_2O/ha/y	0.5	Not fertilized	[50]	

SI.4 Management of agricultural and biomass conversion residuals

The removal of straw from fields induces changes in the soil carbon stock [48, 51-53]. A depletion of 0.3 tonne C/ha of soil organic carbon (SOC) due to straw removal was assumed according to the IPCC carbon tool (Country: Denmark; climate region: cold temperate, moist; native soil type: high clay activity mineral; land use type: long-term cultivated, full tillage, from medium to low input). The average straw yield in Denmark for the years 2006-2008 was 3.246 tonne/ha. As a consequence, the calculated carbon depletion was 0.09 tonne C/tonne straw. Removal of nutrients (N, P and K) with the straw led to additional fertilizer use to maintain constant crops yields. The following values were assumed: 6.5 kg N, 3.4 kg P and 2.8 kg K per tonne DM straw according to [48]. Straw removal also caused lower N_2O emissions: a decrease of 0.03 kgN-N_2O/tonne DM straw was assumed [48].

The use of grass for energy instead of feeding induced an increased demand for other types of fodder. This was modeled with additional production of barley in order to satisfy the feed demand.

The use-on-land of digestate from anaerobic digestion of manure was credited by substitution of inorganic N, P, K fertilizers [15, 16]: 1 tonne of digestate was assumed to substitute 4.07 kg of ammonium nitrate (as N), 2.1 kg of triple superphosphate (as P_2O_5) and 3.3 kg of potassium chloride (as K_2O).

The use-on-land of biochar was also credited for its potential positive effects on soil, e.g. carbon sequestration, improved fertilizer efficiency and reduced N_2O emissions. Based on [54], the content of carbon in the biochar resulting from pyrolysis processes of lignocellulosic materials corresponded to approximately 65% (wt) and the content of stable carbon (i.e. carbon with a residence time in the soil higher than 1000 years) 54% (wt). In gasification processes the carbon content in biochar is much lower due to the higher process temperatures. Carbon conversion efficiencies (CCE) of approximately 80% was reported for lignocellulosic biomass by [10]. The remaining 20% was then found in the biochar. Based on these data, the content of carbon and stable carbon in the biochar from gasification of lignocellulosic was estimated to respectively 45% and 36% (wt). This led to a sequestration of carbon with the biochar applied on soil equal to 360 kg C/tonne biochar. Based on [54], the application of biochar also caused an improved efficiency (by 7.2%) of the applied fertilizers. This, assuming an application rate of biochar of 5 tonne C/ha (as stable carbon), and assuming a typical application rate (for corn) of 154 kg N (ammonium nitrate), 64 kg P_2O_5 and 94 kg K_2O, resulted in less use of ammonium nitrate (3.6 kg N/tonne biochar), triple superphosphate (1.5 kg P_2O_5/tonne biochar) and potassium chloride (2.2 kg K_2O/tonne biochar). Also, the lower use of fertilizers further led to decreased emission of N_2O by 50% (0.394 kg N-N_2O/tonne biochar) [54].

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Appendix B. Miscellaneous

B.1. Inventory report for modelling direct land use changes of perennial and annual crop in Denmark

Given the size of the inventory report, it is included in a separate file.

B.2. Flow charts of selected biomass to energy technologies

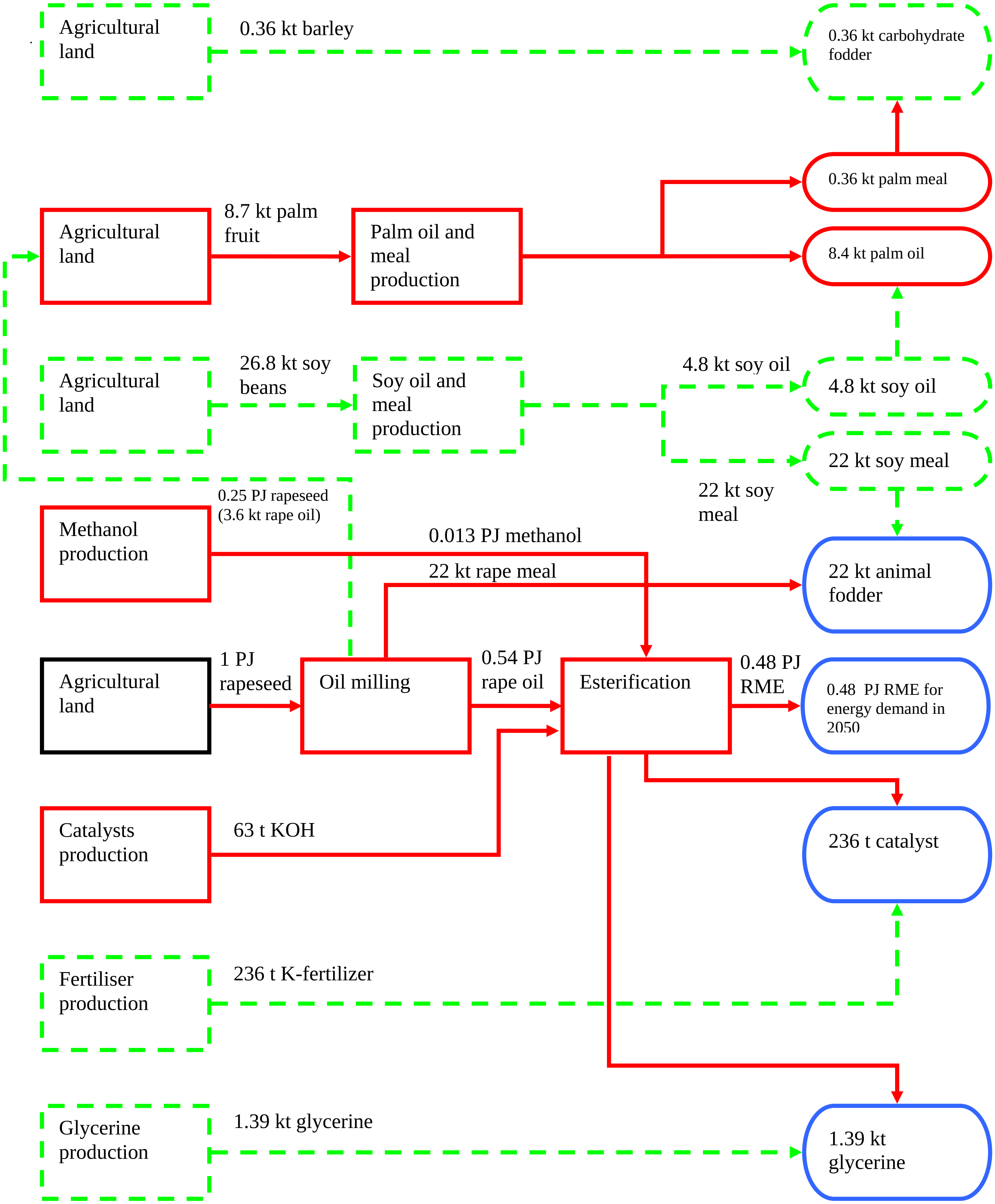

Figure 1 Rapeseed to RME

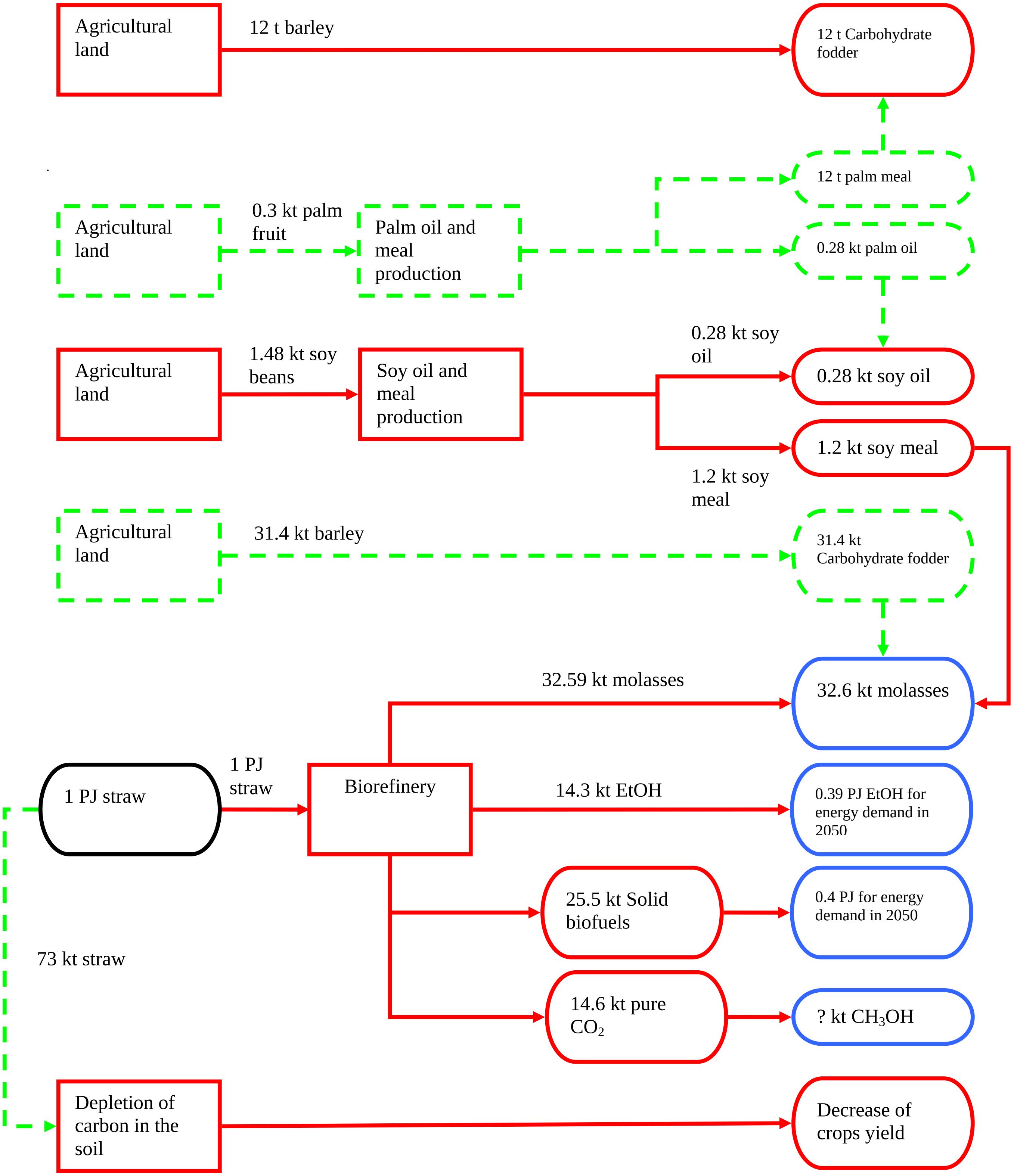

Figure 2 Straw to ethanol

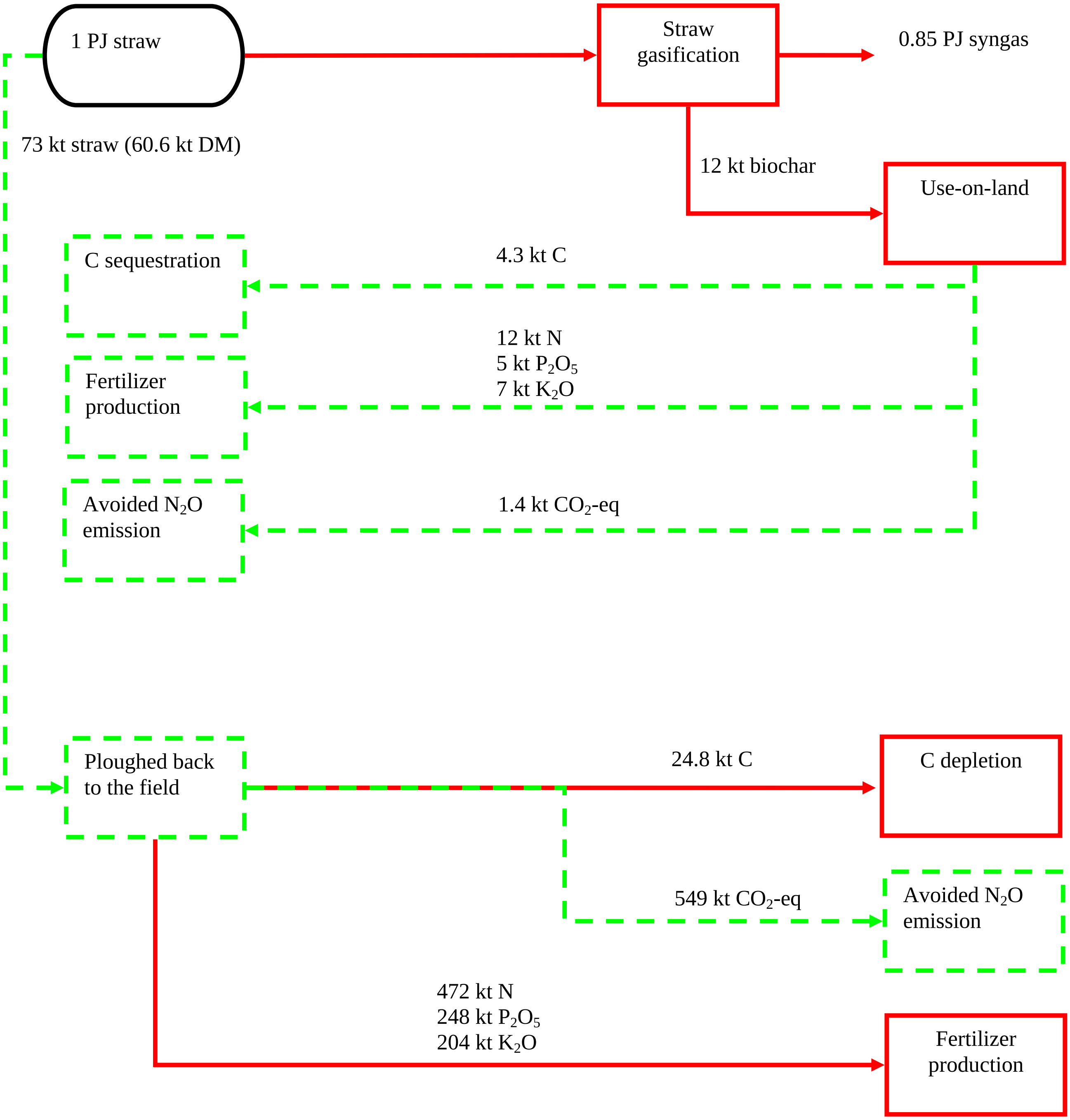

Figure 3 Straw to syngas

The CEESA project (Coherent Energy and Environmental System Analysis) presents technical scenarios as well as implementation policies and a road map of Denmark's transition from a fossil fuel-dominated energy system to a supply system based completely on renewable energy with a dominating part of intermittent sources like wind and solar power. Energy conservation and a certain technological development are prerequisites for this transition. The CEESA scenarios show how the transition can be performed before the year 2050 mainly by the use of known technologies combined with significant energy conservation.

The CEESA project has a focus on, among others, transport, electricity power systems and environmental assessment. The need for new systems thinking and new planning principles for energy investments is among the important observations in this scenario project. With dominant contributions from intermittent sources and limited amounts of biomass available, storage problems are solved by integrating the electricity, heat and transport sectors much more than in traditional supply systems based on fossil fuels. The CEESA project shows how this can be done in an efficient and economical way.

CEESA is a multidisciplinary co-operation which combines the forces of leading Danish researchers in the fields of energy and environment. The project is financed by the Danish Council for Strategic Research together with the participating parties and was conducted in the period 2007-2011.

The results of the CEESA project are presented in 5 background reports and a main summary report.

CEESA main report:

- Coherent Energy and Environmental System Analysis

CEESA background reports:

- Part 1: CEESA 100% Renewable Energy Scenarios towards 2050
- Part 2: CEESA 100% Renewable Energy Transport Scenarios towards 2050
- Part 3: Electric power systems for a transition to 100% renewable energy systems in Denmark before 2050
- Part 4: Policies for a Transition to 100% Renewable Energy Systems in Denmark Before 2050
- Part 5: Environmental Assessment of Renewable Energy Scenarios towards 2050

Aalborg University
Technical University of Denmark
Risø DTU National Laboratory for Sustainable Energy
Pöyry Energy Consulting
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