CARBON MANAGEMENT AND ENVIRONMENTAL CONSEQUENCES OF AGRICULTURAL BIOMASS IN A DANISH RENEWABLE ENERGY STRATEGY

PhD thesis

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PREFACE

This thesis describes the work performed by the author for fulfilling the requirements for a PhD degree in Energy and Environmentally Efficient Technologies at the Institute of Chemical Engineering, Biotechnology and Environmental Technology of University of Southern Denmark (Faculty of Engineering).

The project is affiliated to the 3R • Residual Resources Research graduate school (www.3r.env.dtu.dk) and was made possible through the financial support of the research project Coherent Energy and Environmental System Analysis (CEESA) (www.ceesa.dk), partly financed by The Danish Council for Strategic Research. The overall aim of the multidisciplinary CEESA project was to develop methodologies, present technical scenarios as well as policies leading to a road map for Denmark's transition towards a "100%" renewable energy system. The work described herein was part of a work package within the CEESA project.

The supervision of this PhD work was ensured by Professor Henrik Wenzel of University of Southern Denmark. Significant partners related to this work are PhD student Davide Tonini and assistant professor Thomas Astrup from the Technical University of Denmark, also partners within the CEESA project. Other significant partners include senior scientist Uffe Jørgensen and Professor Jørgen Eivind Olesen from the faculty of agroecology of Aahrus University, as well as Bjørn Molt Petersen, formerly senior scientist at the same institution. Through this active collaboration with Aahrus University, the author has actively participated in other research environments.

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ABSTRACT

The Danish Government has set a long term strategy for Denmark to be independent of fossil fuels in 2050. Several studies have been conducted to design and optimize such a system, and these all highlight the indispensability of a biomass potential of around 35 – 50% of the overall energy consumption, being 300 – 450 PJ y^{-1} of biomass out of Denmark's present 850 PJ y^{-1} overall energy consumption. There are several reasons explaining why biomass is so attractive for energy systems entirely free of fossil energy. Its key advantages, however, lie in the fact that it is storable, and that it is a source of carbon, a resource that becomes limited in a renewable energy future.

Being a country with a relatively high agricultural density, Denmark has access to a considerable amount of agricultural residues that can be used for energy production. However, this potential is still far from enough to supply the biomass requirements of an energy system entirely free of fossil fuels, which involves that conversion of agricultural land from food/feed crops to energy crops is, in Denmark and/or beyond, unavoidable. Further, agricultural residues already have their uses, so using them for energy diverts them from these other applications.

In this context, the overall goal of this study is to provide holistic insights as well as scientific background in determining the environmental consequences of using different agricultural biomasses within a renewable energy system in Denmark. It focuses on the three main agricultural biomass resources available in Denmark: energy crops, harvestable above-ground crop residues (also referred to as "secondary yield", e.g. straw and beet tops) and manure.

The research performed within this PhD work can be divided into three main research lines:

1) Contributing to advances in modeling the environmental consequences of bioenergy production from agricultural biomass (manure, energy crops and crop residues) in Denmark. These advances focus, among others, on the environmental consequences of direct and indirect land use changes;
2) Contributing to advances in consequential life cycle assessment (LCA) modeling involving manure handling;
3) Assessing the environmental consequences of different strategies to boost the manure-biogas production through additional carbon inputs, in the perspective of using 50% of the Danish manure for biogas production by 2020.

In the framework of the first research line, a Danish-specific consequential life cycle dataset for assessing the consequences of direct land use changes caused by energy crops in Denmark has been built. The system boundary included all activities within the cultivation stage (from soil cultivation to harvest) and the reference flow used for each processes was 1 ha of land in a year. A considerable level of details has been included in the inventory, resulting in a total of 528 combinations, comprising 7 crops (annuals and perennials), 2 soil types (sand and sandy loam), 2 climate types (wet and dry), 3 initial soil carbon level (high, average, low), 2 horizon time for soil carbon changes (20 years and 100 years), 2 residues management practices (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 %). For all these combinations, the input and output flows from and to the environment were quantified, including soil carbon changes. Sensitivity analyses were performed with different N fertilizers (manure, urea, calcium ammonium nitrate) as well as with different methodologies for assessing nitrous oxide (N_2O) emissions. Beyond the quantification of the net emission flows related to all combinations considered, key results also included the partition of the dry matter, carbon and nitrogen flows between the crop, the secondary yield, and the above- and below- ground residues of all assessed crop systems.

This life cycle inventory has been used in a bioenergy case study assessing, through consequential LCA, the environmental impacts associated with the production of heat and electricity from one hectare of Danish arable land cultivated with three perennial crops: ryegrass, willow and *Miscanthus*. For each, four conversion pathways were assessed against a fossil fuel reference: i) anaerobic co-digestion with manure, ii) gasification, iii) combustion in small-to-medium scale biomass combined heat and power (CHP) plants and iv) co-firing in large scale coal-fired CHP plants. For each of these 12 bioenergy systems, all relevant carbon and nitrogen flows were disaggregated and quantified for all the major processes involved, including direct and indirect land use changes.

In the framework of the second research line, a methodology was developed in order to handle the challenges of including manure into LCA. The main challenge with manure systems is that the emissions at any point of the system are very much dependent upon the manure composition, which itself undergo several changes throughout the different stages of the system, governed by factors such as the management (e.g. handling, treatment, etc.), or site-specific parameters (e.g. temperature). The essence of the developed methodology consisted of a step-wise procedure to define a reference manure management system, including the establishment of a reference manure composition ex-animal, ex-housing and ex-storage that is consistent with the input and output substance flows to and from the manure continuum. Further, the methodology takes into account the framework conditions laid down by site-specific conditions as well as legislations. The latter was particularly relevant with respect to fertilizer substitution, as it defined the amount of nitrogen substituted by mineral fertilizers, depending on the manure-based product applied on land. Substitution of phosphorus and potassium fertilizers was also addressed by the developed methodology.

The third research line involved two LCA case studies aiming at assessing the environmental consequences of different strategies for supplying a drastic increase of manure-biogas in Denmark. The first case study investigated the possibility of increasing manure-biogas without relying on the availability of external carbon co-substrates. It consisted to co-digest raw pig and cattle slurry together with the concentrated solid fraction resulting from (ex-housing) manure separation. Three scenarios were assessed, each considering different slurry separation technologies to obtain the solid fraction input for biogas production.

In the second case study, additional options were investigated, with a focus on external C co-substrates. Five external co-substrates not already fully used for biogas were considered: energy crops (represented by maize silage), straw, household biowaste, commercial biowaste and garden waste. Further, the use of the solid fraction deriving from source-segregation of animal urine and feces was also investigated. This latter option differed from the first case study, as it involved a separation system directly under the animals, where the contact between feces and urine is prevented at the first place.

The main results of this PhD work can be summarized as follows:

- The market-driven land expansion (i.e. indirect land use change) resulting from using more Danish arable land for energy crop cultivation was shown to offset any potential benefits of bioenergy, unless high-yielding crops (i.e. >10 t DM ha^{-1}) with low water content or low DM losses were considered, in combination with efficient conversion technologies (i.e. 85-90%). The indirect land use changes impact was quantified as 310±170 and 357±195 t CO_2 ha^{-1} displaced, depending on the yield considered for the crop displaced by energy crop in Denmark. Other components of indirect land use changes, like the impacts of intensification, or the foregone sequestration capacity of natural vegetation, were not included.
- All anaerobic co-digestion scenarios highlighted the important environmental benefits, particularly with respect to global warming, of avoiding the reference manure management to take place, i.e. its conventional storage and land application without further processing. As a result, important additional benefits were obtained for the scenarios allowing to use more manure for co-digestion.

This finding also emphasized that manure, in a Danish renewable energy system, should be prioritized for biogas.

- The environmental benefits of using separated solid manure (ex-housing) as a co-substrate to boost the methane production of raw slurry were highly dependent upon the efficiency of the separation technology used to concentrate the volatile solids in the solid fraction. Yet, this biogas production concept appeared as limited in the perspective of a wide-spread strategy for increased manure-biogas. In fact, acknowledging that global warming is a key concern, only one of the studied alternatives allowed for clear GHG reductions compared to the reference slurry management. Yet, this alternative involved the use of a polymer, namely cationic polyacrylamide (PAM), which likely persists and accumulates in the environment, and which does represent a potential toxicity risk, although this could not be quantified in the LCA. On this basis, further research on efficient separation technologies not involving cationic PAM appears necessary.
- Source-segregated solid manure (i.e. obtained from preventing any contact between urine and feces) was highlighted as the co-substrate yielding the greatest environmental benefits overall. This mostly reflected that it allowed to use a lot more manure for biogas than the other scenarios. Although this scenario appeared to be favorable for the long-term, it may not be so realistic to rely on this carbon co-substrate in the short-term perspective (2020), as it would involve major changes and investments in current farm buildings.
- Straw and biowastes (i.e. garden waste as well as household and commercial food waste) should be prioritized for manure-biogas, rather than for their other potential uses (i.e. thermal energy recovery and composting). The rationale for this is that the use of these co-substrates for biogas:
    - Resulted in a lower global warming potential than their use for incineration and composting, respectively;
    - Allowed to recycle these co-substrate's nutrients, including the slowly degradable carbon, which are essentially lost in the incineration case;
    - Produces a storable gas that can be used for both CHP and transport, a key flexibility asset for a renewable energy system involving more than 50% wind power;
    - Allows to avoid energy crops to be used for manure-biogas, and thereby the indirect land use changes related to it.
- Energy crops, because of the land use change they generate, should be used as a last priority within a renewable energy system. However, to the extent they are needed, long duration perennial crops (i.e. *Miscanthus* and willow) should be favored. Particularly, *Miscanthus* was highlighted as the most promising of the investigated energy crops, as it has a relatively high yield, the lowest emission flows of nitrogen compounds, involves relatively low losses of N and P towards aquatic recipients, and allows to increase soil organic carbon. Results however showed that the magnitude of these benefits depends on the harvest season, soil types and climatic conditions.
- Winter wheat was highlighted as the only annual crop where straw removal for bioenergy may be suitable, being the only annual crop not involving losses of soil organic carbon as a result of harvesting the straw. This, however, was only true for sandy soils, and was conditional to manure application. On this basis, and in the light of on-going work on assessing the quality of Danish soils, straw removal should preferably take place on soils with low clay-to-soil organic matter ratio (i.e. <10). Such soils cover most of Jutland, but are also found on Funen and Zealand.
- Finally, it was pointed out that, in a renewable energy future, biomass will become the main source of carbon. In this respect, it was emphasized that carbon efficiency of future biomass & technology combinations will be a decisive concern in a fossil free society.

This PhD work is a platform for a myriad of further research work. First, all case studies performed within this work assumed that bioenergy would displace fossil-fuel based heat and electricity. In the light of the recent energy agreement of the Danish Government, however, it becomes clear that the future energy displaced will most likely consists of a mix of wind and biomass. Such marginal biomass fuel and the environmental consequences it involves should, in future work, be defined. Further, it would be relevant to perform additional case studies considering the interactions with the transport sector. Moreover, this PhD work pinpointed the necessity of additional work on indirect land use changes. Particularly, the land expansion and intensification response resulting from the displacement of 1 ha of Danish land should be assessed through general or partial equilibrium models, and this for a variety of marginal crop displaced from Denmark. This would be an improvement of the rougher approach used in this study to address indirect land use changes, and could subsequently be used, in combination with the direct land use change inventory developed herein, for modeling any direct and indirect land use changes from increased energy crops in Denmark. Future framework conditions, like a tremendous increase of bioenergy worldwide, drastic changes in the amount and type of food/feed demanded worldwide, phosphorus or pollinator limitations should similarly be addressed. Moreover, additional strategies for increasing the methane produced from manure biogas could be investigated, like for example through the use of recycled carbon dioxide from stationary combustion processes and hydrogen from the electrolysis of water, powered by surplus electricity (e.g. wind).

RESUMÉ

Den danske regering har lagt en langsigtet strategi, hvor målet er at gøre Danmark uafhængigt af fossile brændsler i 2050. Flere studier er blevet gennemført for at designe og optimere et sådant system, og alle disse fremhæver det nødvendige i at udnytte biomassepotentialet på omkring 35-50% at det samlede energiforbrug, svarende til 300 – 450 PJ per år baseret på biomasse, ud af Danmarks nuværende energiforbrug på 850 PJ per år. Der er flere grunde til at biomasse er så attraktivt for et energisystem, der er fuldstændigt uafhængigt af fossil energi. De væsentligste fordele ligger i, at det kan opbevares, og at det er en kulstofkilde, en ressource, som vil blive begrænset i et fremtidigt samfund baseret på vedvarende energi.

En relativt stor andel af arealet i Danmark anvendes til landbrug, og Danmark har dermed adgang til væsentlige mængder restprodukter fra landbruget, og disse restprodukter kan anvendes til energiproduktion. Potentialet er imidlertid langt fra tilstrækkeligt til at kunne forsyne den efterspørgsel efter biomasse, der vil være i et samfund baseret udelukkende på vedvarende energikilder, hvilket uundgåeligt vil medføre at landbrugsarealer, der anvendes til fødevarer- eller foderafgrøder, skal konverteres til energiafgrøder i Danmark og/ eller i udlandet. Desuden udnyttes restprodukterne fra landbruget allerede, og hvis restprodukterne udnyttes til energiproduktion, vil de ikke være tilgængelige for de nuværende formål.

I denne sammenhæng er det overordnede mål med nærværende afhandling at give en helhedsorienteret indsigt såvel som den videnskabelige baggrund for at kunne fastlægge de miljømæssige konsekvenser af at udnytte forskellige biomasser fra landbruget i et energisystem baseret på vedvarende energi i Danmark. Afhandlingen fokuserer på 3 primære biomasseressourcer fra landbruget, som er tilgængelige i Danmark: Energiafgrøder, høstbare overjordiske afgrøderester (også kaldet ”det sekundære udbytte”, f.eks. halm og roetoppe) samt husdyrgødning.

Den forskning, der er gennemført med denne PhD afhandling, kan opdeles i tre forskningsområder:

1) At bidrage til udviklingen af modelleringen af de miljømæssige konsekvenser af energiproduktion ud fra biomasse fra landbruget (husdyrgødning, energiafgrøder og restprodukter fra afgrøder) i Danmark. Denne udvikling fokuserer blandt andet på de miljømæssige konsekvenser af direkte og indirekte ændringer i arealudnyttelsen.
2) At bidrage til udviklingen af metoderne for modellering af livscyklusvurderinger (LCA), baserede på konsekvenstankegangen, med fokus på modellering af håndtering af husdyrgødning.
3) At vurdere de miljømæssige konsekvenser af forskellige strategier for at styrke biogasproduktion baseret på husdyrgødning via ekstra tilførsel af kulstof, i perspektivet af at udnytte 50% af den danske husdyrgødning til biogasproduktion i 2020.

Inden for rammerne af det første forskningsområde, er der opbygget et specifikt dansk datasæt for konsekvens-LCA til at vurdere konsekvenserne af de ændringer, energiafgrøder i Danmark har for direkte arealudnyttelse. Systemgrænserne inkluderede alle aktiviteter i dyrkningsfaserne (fra jordbearbejdning til høst). Referencen for hver proces var 1 ha areal i ét år. Detaljeringsniveauet i opgørelsen er betydeligt, og omfatter 528 kombinationer, 7 afgrøder (ét-årige og flerårige planter), 2 jordbundstyper (sand samt sandholdig jordler), 2 klimatyper (våd og tør), 3 niveauer af kulstofindholdet i jorden (højt, gennemsnitligt og lavt), 2 tidshorisonter for udviklingen i jordens indhold af kulstof (20 år og 100 år), 2 scenarier for håndtering af afgrøderester (fjernelse og indarbejdelse i jorden) såvel som 3 niveauer for reduktion af omsætningen af jordens kulstofindhold for flerårige planter som konsekvens af fraværet af jordbearbejdning (0, 25 %, 50 %). For alle disse kombinationer blev de indgående og udgående strømme kvantificeret, inklusive ændringer i jordens indhold af kulstof. Følsomhedsanalyser blev gennemført med forskellige N-holdige gødningstyper (husdyrgødning, urea, calcium ammonium nitrate) så vel som forskellige metoder til vurdering af lattergasemissioner (N_2O). Udover at kvantificere de netto-emissioner, der var relateret til alle de involverede

kombinationer, inkluderede resultaterne også opdeling af tørstof, kulstof og nitrogen mellem afgrøde, det sekundære udbytte samt de høstbare over- og underjordiske jordiske afgrøderester for alle de inkluderede systemer af afgrøder.

Denne livscyklusopgørelse blev anvendt i et bioenergy *case studie*, hvor de miljømæssige påvirkninger i forbindelse med produktionen af varme og elektricitet fra én hektar dansk landbrugsjord dyrket med tre flerårige afgrøder: rajgræs, pil og elefantgræs. Vurderingerne blev gennemført ved hjælp af konsekvens-LCA. For hver afgrøde blev fire scenarier for udnyttelse af afgrøden vurderet: i) anaerob omsætning sammen med husdyrgødning ii) forgasning, iii) forbrænding i et biomassebaseret anlæg der producerer kombineret kraft-varme (skala: lille til medium størrelse anlæg) iv) forbrænding sammen med kul i et storskala kulfyret kraftvarmeværk. For hvert af disse 12 bioenergisystemer blev alle relevante strømme af kulstof og nitrogen disaggregeret og kvantificeret for alle de væsentligste processer, der var involveret, inklusiv ændringerne i direkte og indirekte arealanvendelse.

Inden for rammerne af det andet forskningsområde blev en metode udviklet med det formål at kunne håndtere de udfordringer, der ligger i at inkludere husdyrgødning i LCA. Den største udfordring med husdyrgødning er, at emissionerne et hvert sted i systemet er afhængige af husdyrgødningens sammensætning, som selv gennemgår konstante forandringer gennem de forskellige stadier i systemet, der i endvidere er reguleret af faktorer som håndtering og behandling samt stedspecifikke faktorer (f.eks. temperatur). Kernepunktet i den udviklede metode bestod af en trinvis procedure for at definere et referencesystem for husdyrsgødningshåndteringen. Dette inkluderede etableringen af en referencesammensætning for husdyrgødningen ab dyr, ab stald og ab lager, som var konsistent med de indgående og udgående flows til og fra husdyrgødningen. Endvidere inkluderer metoden stedspecifikke forhold så vel som lovgivning. Sidstnævnte har især været relevant med hensyn til gødningssubstitution, fordi lovgivningen definerede den mængde nitrogen i kunstgødning, som nitrogen i husdyrgødning erstatter, og som afhænger af den type husdyrgødning, som spredes på marken. Fosfor og kalium blev også inkluderet i metoden.

Det tredje forskningsområde involverede to LCA *case studier* med det formål at vurdere de miljømæssige konsekvenser af forskellige strategier for en signifikant forøgelse af biogasproduktion baseret på husdyrgødning i Danmark. Det første studie undersøgte muligheden for en forøgelse af biogasproduktionen baseret på husdyrgødning uden at være afhængigt af tilgængeligheden af eksterne kulstofkilder som co-substrat. Det første *case studie* bestod i at samkøre rågylle fra svin og kvæg med den opkoncentrerede faste fraktion, der opstår, hvis man separerer husdyrgødning (ab stald). Tre scenarier blev vurderet, hver med forskellige gylle-separeringsteknologier for at gøre den faste fraktion tilgængelig for biogasproduktion.

I det andet *case studie* blev yderligere muligheder undersøgt med fokus på eksterne kulstof co-substrater. Undersøgelsen omfattede fem eksterne co-substrater, som ikke allerede anvendes til biogasproduktion i fuld udstrækning: energiafgrøder (repræsenteret ved majssilage), halm, bioaffald fra husholdninger, kommercielt bioaffald samt haveaffald. Endvidere blev udnyttelsen af den faste fraktion fra kilde-separering af husdyrgødningen (urin og fæces) undersøgt. Sidstnævnte mulighed adskiller sig fra det første *case studie* ved at separationen foregår direkte under dyrene i stalden, hvor kontakt mellem urin og fæces forhindres fra begyndelsen.

Hovedresultaterne af nærværende PhD-afhandling kan opsummeres som følger:

- Den markedsdrevne areaudvidelse (f.eks. indirekte ændringer i arealanvendelse) som resultat af at anvende mere Dansk landbrugsjord til dyrkning af energiafgrøder viste sig at opveje alle potentielle fordele ved bioenergi, medmindre meget effektive konverterings-teknologier blev anvendt (dvs. 85-90%), i kombination med afgrøder med højt udbytte (dvs. >10 t tørstof per ha). Virkningen af de indirekte ændringer i arealanvendelsen blev kvantificeret til 310±170 and 357±195 ton CO_2 ha^{-1} fortrængt, afhængig af udbyttet for den fortrængte afgrøde (fortrængt af energiafgrøde i Danmark).

Øvrige komponenter af indirekte ændringer i arealanvendelsen, som virkninger af intensivering, eller den mistede sekvestreringskapacitet af naturlig vegetation blev ikke inkluderet.

- Alle anaerobe *co-digestion* scenarier fremhævede de vigtige miljømæssige fordele, især med hensyn til den globale opvarmning, for at undgå referencescenariets gødningshåndtering at finde sted, dvs. den konventionelle lagring og tilførsel af husdyrgødning til jorden uden yderligere forarbejdning. Som resultat af dette, kunne yderligere fordele opnås for de scenarier, der involverede en øget anvendelse af husdyrgødning til *co-digestion*. Denne konklusion understreger endvidere, at det bør prioriteres at anvende husdyrgødning til biogasproduktion i et dansk energisystem baseret på vedvarende energi.
- De miljømæssige fordele ved at anvende den faste del af separeret husdyrgødning (ab stald) som et co-substrat for at øge metanproduktionen af rå-gylle var meget afhængig af effektiviteten af den separationsteknologi, der blev anvendt til at separere og opkoncentrere de omsættelige organiske forbindelser ("volatile solids", VS) i den faste fraktion. Ikke desto mindre forekom dette koncept for biogasproduktion at være begrænset i perspektivet af en øget anvendelse af biogas produceret på dyregødning. I erkendelsen af, at den globale opvarmning er et meget centralt miljøproblem, var der faktisk kun ét af de undersøgte alternativer, der reducerede drivhusgasemissionerne sammenlignet med referencescenariet. Men dette alternativ involverer anvendelsen af en polymer, nemlig kationisk polyacrylamid (PAM), der sandsynligvis akkumuleres i miljøet, og som udgør en potentiel toksisk risiko, endskønt dette ikke kunne kvantificeres i LCA'en. På dette grundlag forekommer yderligere forskning i effektive separationsteknologier, der ikke involverer kationisk PAM, nødvendigt.
- Kilde-separeret fast gødning (dvs. fremstillet ved at forhindre enhver kontakt mellem urin og fæces) blev fremhævet som det co-substrat, der giver de største miljømæssige fordele samlet set. Dette afspejlede især, at dette scenarie muliggjorde anvendelse af væsentligt større mængder husdyrgødning til biogas, end de andre scenarier. Selv om denne situation viste sig at være gunstig på lang sigt, vil det ikke være realistisk at basere sig på dette kulstof co-substrat på kort sigt (2020), da det ville medføre store ændringer og investeringer i de nuværende landbrugsbygninger.
- Det bør prioriteres at anvende halm og bioaffald (dvs. haveaffald samt bioaffald fra husholdninger og kommercielt madaffald) til biogasproduktion, fremfor de andre potentielle anvendelsesmuligheder (dvs. termisk energiudnyttelse og kompostering). Begrundelsen for dette er, at anvendelsen af disse co-substrater for biogas:
    - Resulterede i et lavere bidrag til global opvarmning end deres anvendelse til henholdsvis forbrænding og kompostering;
    - Giver mulighed for at genbruge disse co-substraters næringsstoffer, herunder tungt nedbrydelige kulstofforbindelser, som i mistes ved forbrænding.
    - Giver en gas, der kan lagres, og som kan anvendes både til kraftvarme og transport, en central fleksibilitet, som er et aktiv for et energisystem baseret på vedvarende energi, og som involverer mere end 50% vindkraft;
    - Giver mulighed for at undgå at energiafgrøder skal anvendes til biogas, og dermed de indirekte ændringer i arealanvendelsen i forbindelse med det.
- Energiafgrøder bør anvendes som en sidste prioritet i et energisystem baseret på vedvarende energi på grund af de ændringer i arealanvendelsen, de genererer. Men i det omfang, de er nødvendige, bør flerårige afgrøder med lang livscyklus (dvs. elefantgræs og pil) favoriseres. Især blev elefantgræs bør fremhæves som den mest lovende af de undersøgte energiafgrøder, da det har et relativt højt udbytte, de laveste emissioner af kvælstofforbindelser, involverer relativt lave tab af N og P til akvatiske recipienter, og gør det muligt at øge jordens indhold af organisk kulstof. Resultaterne viste imidlertid, at omfanget af disse fordele afhænger af høstsæsonen, jordtyper og klimaforhold.
- Vinterhvede blev fremhævet som den eneste årlige afgrøde, hvor fjernelse af halm til anvendelse som bioenergi kan være egnet, da det er den eneste ét-årlige afgrøde, som ikke indebærer tab af organisk kulstof som et resultat af at høste strå. Dette er dog kun var tilfældet for sandholdige jorder, og var betinget af tilførsel af husdyrgødning. På dette grundlag og på baggrund af det igangværende

arbejde med at vurdere kvaliteten af danske jorde, bør fjernelse af halm fortrinsvis finde sted på jorde med lav ler-til-jord organisk stof forhold (dvs. <10). Sådanne jordtyper dækker det meste af Jylland, men findes også på Fyn og Sjælland

- Endelig blev det fremhævet, at biomasse vil blive den vigtigste kulstofkilde i et fremtidigt samfund baseret på vedvarende energi. I denne forbindelse blev det understreget, at kulstofeffektivitet af fremtidige kombinationer af biomasse & teknologi vil være et afgørende hensyn i et samfund, der er uafhængigt af fossile energikilder.

Denne PhD afhandling er en platform for et utal af yderligere forskningsarbejde. I alle *case studier* gennemført i denne afhandling er det forudsat / antaget, at bioenergi vil fortrænge varme og elektricitet baseret på fossile brændsler. I lyset af den seneste energipolitiske aftale indgået af den danske regering, står det dog klart, at fremtidens fortrængte energi sandsynligvis vil bestå af en blanding af vind og biomasse. En sådan marginal biomassebrændsel og de miljømæssige konsekvenser af, hvad det indebærer, bør defineres i fremtidigt arbejde. Desuden vil det være relevant at udføre yderligere *case studier*, hvor interaktioner med transportsektoren tages med i betragtning. Desuden peger denne PhD-afhandling på nødvendigheden af yderligere arbejde med indirekte ændringer i arealanvendelsen. Især bør areal ekspansion og intensivering som følge af fortrængning af 1 ha dansk jord vurderes ved generelle eller partielle ligevægtsmodeller og dette bør foretages for en række marginale afgrøder fortrængt fra dansk areal. Det vil være en forbedring af den grovere tilgang, der anvendes i denne afhandling for at adressere de indirekte ændringer i arealanvendelsen, og det vil efterfølgende kunne anvendes i kombination med den opgørelse af direkte ændringer i arealanvendelsen, som er udviklet i nærværende afhandling, til at modellere alle direkte og indirekte ændringer i arealanvendelsen fra en øget anvendelse af energiafgrøder i Danmark. Fremtidige rammebetingelser, som en enorm forøgelse af bioenergi på verdensplan, drastiske ændringer i efterspørgselen af mængden og typen af fødevarer/foder hele verden, fosfor eller ændringer i insektbestøvning bør ligeledes tages op. Endvidere kunne yderligere strategier til at øge metan produceret fra biogas baseret på husdyrgødning undersøges, som for eksempel gennem brug af genanvendt kuldioxid fra stationære forbrændingsprocesser og brint fra elektrolyse af vand, drevet af overskydende elektricitet (f.eks vind).

TABLE OF CONTENT

LIST OF PAPERS

The thesis is based on the following papers:

Paper *I*: Hamelin L, Jørgensen U, Petersen BM, Olesen JE, Wenzel H (2012). Modelling the carbon and nitrogen balances of direct land use changes from energy crops in Denmark; a consequential life cycle inventory. *Global Change Biology Bioenergy*, 4, 889-907.

Paper *II*: Tonini D, **Hamelin L**, Wenzel H, Astrup T (2012). Bioenergy production from perennial energy crops: a consequential LCA of 12 bioenergy chains including land use changes. *Environmental Science & Technology,* 46, 13521-13530.

Paper *III*: Hamelin L, Wenzel H (2011). Methodological aspects of environmental assessment of livestock production by LCA (Life Cycle Assessment). In: *Emissionen der Tierhaltung. Treibhausgase, Umweltbewertung, Stand der Technik* (eds KTBL), p. 34-53. Darmstadt, Germany.

Paper *IV*: Hamelin L, Wesnæs M, Wenzel H, Petersen BM (2011). Environmental consequences of future biogas technologies based on separated slurry. *Environmental Science & Technology*, 45, 5869-5877.

Paper *V*: Hamelin L, Naroznova I, Wenzel H. Environmental consequences of different carbon alternatives for increased manure-based biogas. *To be submitted.*

The author's contribution to the above-mentioned papers is detailed in the co-author statements submitted along with this thesis. In a nutshell, the author's contribution can be summarized as follows:

Paper ***I***: The author performed ca. 70% of the modeling work, and 100% of the article's redaction. Reviewer's comments were addressed entirely by the author.

Paper ***II***: The concept of the paper was fully established by the author and co-author D. Tonini. The modeling work was performed in the proportion of ca. 60% by D. Tonini, and 40% by the author. The author's contribution to the writing of the article corresponds to more than 60% of the share. Reviewer's comments were addressed in equal share by L. Hamelin and D. Tonini.

Paper ***III***: Overall, the contribution of the author is estimated to ca. 60%, and co-author Henrik Wenzel contributed to ca. 40%.

Paper ***IV***: The author participated to the modeling work in equal share with co-author M. Wesnæs (ca. 45% each), and contributed to 100% of the article's redaction. Reviewer's comments were addressed entirely by the author.

Paper ***V***: The author contributed to ca. 40% of the modeling work, and 100% of the article's redaction.

Papers ***I***, ***II*** and ***IV*** are published in scientific journals, while paper ***V*** will be submitted to a scientific journal in early 2013. Paper ***III*** is a peer-reviewed conference paper (invited paper), but will be slightly adapted and submitted to a scientific journal as well.

RELATED PUBLICATIONS NOT INCLUDED IN THIS THESIS

Peer-reviewed scientific journals

1. De Vries JW, Vinken TMWJ, **Hamelin L**, De Boer IJM (2012). Comparing environmental consequences of anaerobic mono-and co-digestion alternatives for pig manure to produce bio-energy – a life cycle perspective. *Bioresource Technology*, 125, 239-248.

Peer-reviewed reports

2. **Hamelin L,** Wesnæs M, Petersen BM, Wenzel H (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
3. Lund H, Hvelplund F, Mathiesen BV, Østergaard PA, Christiensen P, Connolly D, Schaltz E, Pillay JR, Nielsen MP, Felby C, Bentsen NS, Meyer NI, Tonini D, Astrup T, Heussen K, Morthorst PE, Andersen FM, Münster M, Hansen L-LP, Wenzel H, **Hamelin L**, Munksgaard J, Karnøe P, Lind M (2011). Coherent energy and environmental system analysis, a strategic research project financed by the Danish Council for strategic research, programme commission on sustainable energy and environment. Department of Development and Planning, Aalborg University, Aalborg, Denmark; http://www.ceesa.plan.aau.dk/digitalAssets/32/32603_ceesa_final_report_samlet_02112011.pdf

Non peer-reviewed reports

4. **Hamelin L** (2011). Inventory report for modelling direct land use changes of perennial and annual crops in Denmark. Version 0, Presented for the CEESA WP5 report. http://www.ceesa.plan.aau.dk/digitalAssets/24/24178_lci-report---direct-luc-data-for-selected-e-crops-v18-09-11-2010-ceesa.pdf
5. **Hamelin L**, Wenzel H (2012). Analytical framework for assessing techniques for the revision of the BREF document on intensive rearing of poultry and pigs. Submitted to the European IPPC Bureau, Seville.

Articles in Danish

6. Mathiesen VB, Lund H, Hvelplund FK, Connolly D, Bentsen NS, Tonini D, Morthorst PE, Wenzel H, Astrup T, Meyer NI, Münster M, Østergaard PA, Bak-Jensen B, Nielsen MP, Schaltz E, Pillai JR, **Hamelin L**, Felby C, Heussen K, Karnøe P, Munskgaard J, Pade L-L, Andersen FM (2012). Det intelligente energisystem, Biomasse I fremtidens danske energisystem, Robust og bæredygtig bioenergy, 30-31.

Peer-reviewed conference papers

7. **Hamelin L**, Tonini D, Astrup T, Wenzel H (2012). Bioenergy production from perennial energy crops: a consequential LCA of 12 bioenergy chains including land use changes. In: *Corson MS & van der Werf HMG (Eds), Proceedings of the 8th International Conference on Life Cycle Assessment in the Agri-Food sector (LCA Food 2012)*, 1-4 October 2012, Saint-Malo, France. INRA, Rennes, France, p. 239-244.
8. **Hamelin L**, Wenzel H (2012). Methodological aspects of environmental assessment of livestock production by LCA (Life Cycle Assessment). *2012 IX International Livestock Environment Symposium (ILES IX), ILES-12-094*5. Available on ASABE website, American Society of Agricultural and Biological Engineers: http://elibrary.asabe.org/abstract.asp?aid=41541&t=2&redir=&redirType=

Conference abstracts

9. **Hamelin L**, Jørgensen U, Petersen BM, Olesen JE, Wenzel H (2011). Modelling environmental consequences of direct land use changes from energy crops in a self-sustained and fully renewable energy system in Denmark: Effect of crop types, soil, climate, residues management, initial carbon level and turnover time. Quantifying and managing land use effects of bioenergy, IEA Bioenergy tasks 38, 40 and 43, Campinas, Brazil. Pp. 41. (presented as an oral presentation)
10. **Hamelin L**, Petersen BM, Jørgensen U, Olesen JE, Wenzel H (2010). Towards a 100% renewable energy system in Denmark in 2050: a consequential based LCI for selected energy crops. LCAfood 2010. (presented as a poster).
11. **Hamelin L,** Wesnæs M, Wenzel H, Petersen BM (2010). Anaerobic digestion as a slurry management strategy: a consequential life cycle assessment. Paper CSBE101132, XVIIth World Congress of the International Commission of Agricultural and Biosystems Engineering (CIGR). (presented as an oral presentation)

APPENDIXES

- Appendix A: Supporting Information to Paper ***I***, as available online
- Appendix B: Supporting Information to Paper ***II***, as available online
- Appendix C: Supporting Information to Paper ***IV***, as available online
- Appendix D: Supporting Information to Paper ***V***, (to be submitted)

ABBREVIATIONS AND NOTATION

This section lists some of the most commonly used abbreviations and notation symbols throughout this PhD work.

AG	Above-ground
BG	Below-ground
BtE	Biomass-to-energy
C	Carbon
CAN	Calcium ammonium nitrate
CCE	Carbon conversion efficiency (for syngas)
CGE	Cold gas efficiency (for syngas)
CHP	Combined heat and power
CH_4	Methane
CO_2	Carbon dioxide
CRF	Cumulative radiative forcing
Cu	Copper
DAP	Diammonium phosphate
DLUC	Direct land use change
DM	Dry matter
FM	Fresh matter
FU	Functional unit
GHG	Greenhouse gases
GWP	Global warming potential (metric for the global warming impact category)
H_2	Hydrogen
ILUC	Indirect land use changes
K	Potassium
K_2O	Potassium oxide
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LHV	Lower heating value
LUC	Land use changes
N	Nitrogen
NH_3	Ammonia
NMVOC	Non-methane volatile organic compound
NO	Nitric oxide
NO_x	Nitrogen oxides (NO + NO_2)
NO_2	Nitrogen dioxide
NO_3^-	Nitrate
N_2	Nitrogen gas
N_2O	Nitrous oxide
P	Potassium
P_2O_5	Phosphorus pentoxide
RED	Renewable energy directive
SI	Supporting information
TAN	Total ammoniacal nitrogen
TH	Time horizon
TS	Total solids
UES	Unprotected ecosystem equivalent (metric for the acidification impact category)
VS	Volatile solids
Zn	Zinc

1. INTRODUCTION

1.1 Biomass in a renewable energy strategy

Whether it is for mitigating climate change, ensuring rural income or for fostering the security of energy supply, there is an increasing interest worldwide towards the development of renewable energy systems. In this context, several countries developed targets and legislations endeavoring to reduce their reliance upon fossil fuels (GBEP/FAO 2008).

In Denmark, the Danish Government has set a long term strategy to be independent of fossil fuels in 2050 (Danish Government 2011). This involves that Denmark's entire energy supply, whether for electricity, heat or transport, is to be covered by renewable energy. Several studies have been conducted to design and optimize such a system (Lund *et al.* 2011; Danish Commission on Climate Change Policy 2010; Energinet.dk 2010). These studies all point to the need for a biomass potential of around 35 – 50% of the overall energy consumption, being 300 – 450 PJ y^{-1} of biomass out of Denmark's present 850 PJ y^{-1} overall energy consumption.

There are, as detailed in Wenzel (2010), several reasons explaining why biomass is so attractive in a renewable energy system. On the one hand, it has the key advantage of being storable, and as such can ensure flexibility in balancing the fluctuating energy production from intermittent sources like wind, solar and wave power. Moreover, it can be used for producing high energy density fuels needed for mobility purposes. This applies especially for aviation fuels for which there are likely no other alternatives, but also for sea and long distance road transport, both of which are highly dependent upon energy dense fuels in order to save space and weight for the transport of goods. Finally, biomass represents a source of C, which is at the core of most materials and chemicals consumed nowadays. The C needed for producing these is presently provided mainly by fossil fuels, but in a renewable energy system, biomass becomes the main supplier of C. On the perspective of a transition towards a fully renewable energy system, biomass also has the advantage that it can contribute to electricity, heat and transport energy requirements with relatively minor technical adaptations.

In recent studies, the residual biomass (i.e. excluding energy crops) potential available in Denmark has been estimated to 150 – 200 PJ y^{-1} (Danish Commission on Climate Change Policy, 2010; Mathiesen *et al.* 2009; Energinet.dk 2010; Tonini & Astrup 2012; Gylling *et al.* 2012). This considerable amount of residues, representing ca. 20% of Denmark's overall energy consumption, reflects that Denmark is a country with a relatively high agricultural density. In comparison, the available EU residual biomass potential is, based on the data from Panoutsou *et al.* (2009) and BP (2012), estimated to ca. 10% of the current EU primary energy consumption.

But even in spite of this, the residues potential (150 – 200 PJ y^{-1}) is still not enough to supply the biomass required to satisfy all demands in the fossil free system (300 – 450 PJ y^{-1}). This implies that land-dependent biomass (e.g. energy crops or forest trees), whether it is cultivated in Denmark or imported, appears, in a Danish renewable energy future, inevitable. Although renewable, land-dependent biomass is not unlimited in supply, and can, to the extent it leads to the conversion of natural ecosystems to agriculture, involve tremendous environmental costs (e.g. Edwards *et al.* 2008; Searchinger *et al.* 2008; Gibbs *et al.* 2008). This is especially true if cultivating more energy crops in Denmark leads to the displacement of food or feed crops and thereby, through market mechanisms, to the conversion of lands rich in carbon content (such as tropical land), elsewhere in the world. For example, tropical deforestation, which has so far been essentially driven by agricultural expansion, was estimated, at the end of the twentieth century, to account for ca. 20% of the annual worldwide CO_2 emissions (IPCC 2007).

1.2 Aim of the project

The premise of this study is that agricultural biomass is a core resource for a renewable energy strategy such as the one Denmark aims for. To ensure that the inclusion of this resource in such strategy remains within the boundaries of sustainability, integrated schemes considering both the agriculture and energy sectors are needed. The overall goal of this study is to provide holistic insights as well as scientific background in determining the environmental consequences of using different agricultural biomasses within a renewable energy system in Denmark.

This PhD work thus contributes to advances in modeling the environmental consequences of bioenergy production from agricultural biomass. It focuses on the three main agricultural biomass resources available in Denmark: energy crops, harvestable above-ground crop residues (also referred to as “secondary yield”, e.g. straw and beet tops) and manure.

Specific objectives are:

1) To develop tools and methodologies for modeling the direct land use changes resulting from increases in bioenergy production in Denmark, with emphasis on the changes in N and C flows. This shall include soil C changes and allows to address the issue of using harvestable crop residues for bioenergy production (Hamelin *et al.* ***I***).

2) To apply the methodologies/tools developed in 1) to a relevant bioenergy case study for Denmark (Tonini *et al.* ***II***). This case study should also address the environmental consequences of indirect land use changes.

3) To develop methodologies for considering manure into life cycle assessments, allowing to tackle the dependency of emissions upon manure composition throughout the different stages of the manure system (Hamelin & Wenzel ***III***).

4) Based on the methodologies developed in 3), to assess the environmental consequences of different strategies to boost Denmark’s manure-biogas production through external carbon inputs (Hamelin *et al.* ***IV & V***).

The core of this PhD thesis, where these specific objectives are addressed, consists of the 5 above-mentioned papers, found at the end of this thesis. Sections 2 to 5 of the thesis provide in-depth support to the modeling choices performed in these papers as well as explanations for most theoretical concepts covering the practice of biosystems LCA in general. The description of the main methodologies used in this PhD work is presented in sections 6 to 9. Sections 10 and 11 provide a discussion and a conclusion, respectively, of papers ***I** to **V**’* main findings. The “Supporting Information” (SI) material supporting papers ***I***, ***II***, ***IV*** and ***V*** is available in Appendix A to D, respectively. The mass balances and calculations behind the results presented in these papers are transparently detailed in this supporting information material.

2. STATE-OF-THE-ART CONSEQUENTIAL LCA MODELING

2.1 Life cycle assessment

This section intends to give a short introduction to the life cycle assessment (LCA) methodology in general, as this methodology was applied throughout this PhD work. In brief, LCA is a standardized comparative environmental assessment methodology (ISO, 2006a;b) which consists of assessing and comparing the environmental impacts of selected product/service alternatives from “cradle-to-grave”, i.e. from raw materials extraction, through processing and product manufacturing, encompassing product use and maintenance and finally including ultimate disposal of the product at the end of its lifetime. A state-of-the-art LCA thus includes all significant flows from and to the environment involved in the studied system[1], aggregates them over all life cycle stages and subsequently expresses them per unit of function delivered by the system in question, which in LCA is referred to as the “functional unit”. This latter stage ensures that the comparison is based on the delivery of the same service in all systems compared. These aggregated substance flows are then related to an impact category and the contribution from each substance to these impact categories is quantified through a procedure known as life cycle impact assessment (LCIA). This procedure, divided in 4 main steps, is extensively described in Hauschild (2005), Hauschild *et al.* (2012) as well as in Pennington *et al.* (2004). It allows, for a given impact category (e.g. global warming), to express the aggregated flows as an indicator, or reference substance, (e.g. kg CO_2 eq.) per functional unit, through the use of equivalence factors (e.g. 25 kg CH_4 per kg CO_2 eq.). Different LCIA methodologies have been developed (Pennington *et al.* 2004; Hauschild *et al.* 2012), and these differ on an number of aspects, such as the reference substances and equivalence factors used[2], as well as the type of impact level addressed (midpoint, endpoint, or both[3]).

All LCAs presented in this PhD work rely on the Danish EDIP2003 method described in Hauschild & Potting (2005). Four main impact categories were considered: global warming (over a 100 y horizon), acidification and nutrient enrichment (distinguishing between N and P being the limiting nutrient for growth), these being seen as the most relevant for agricultural biomass systems (Hamelin & Wenzel, ***III***). In Hamelin *et al.* (***IV***), other impacts were also addressed, like the formation of photochemical ozone (smog) and the emission of particles and dust causing respiratory problems (an impact termed “respiratory inorganics”). The nutrient enrichment impact was, throughout this PhD work, referred to as “eutrophication-N” (when N is the limiting nutrient for growth) and “eutrophication-P” (when P is the limiting nutrient for growth).

Background (or generic) life cycle inventory (LCI) data (e.g. machinery, capital goods, mineral fertilizers production, etc.) were taken from the Ecoinvent v. 2.2 database (Frischknecht & Rebitzer, 2005), which contains more than 4000 thoroughly documented and reviewed LCI datasets covering several activity sectors, including energy and agriculture. All LCAs performed were facilitated with the LCA software SimaPro 7.3.3. Foreground (or system-specific) LCI data used are detailed in Hamelin/Tonini *et al.* (***I, II, III, IV, V***).

Additional details on the general principles of the LCA methodology can be found in the ISO standards (ISO, 2006a;b) as well as in Rebitzer *et al.* 2004. Latest developments are summarized in Finnveden *et al.* (2009), and historical details on the development of the LCA methodology can be encountered in Russell *et al.* (2005).

[1] The compilation of these flows is referred to as the Life Cycle Inventory (LCI).

[2] For global impacts, namely global warming and ozone depletion, most LCIA methodologies use the same equivalence and characterization factors, at least for the midpoint level (Hauschild *et al.* 2012). Global impacts are these impacts whose effects do not depend upon the specific location where the causing emissions take place.

[3] Midpoint levels refer to a first level impact from the emission and its immediate, or early-stage, effect (e.g. overall increased radiative forcing at the surface of Earth, increased UV intensity at Earth’s surface, algal bloom, etc.), while endpoint levels refer to the ultimate consequence of an environmental impact (e.g. loss of human life, loss of crops, loss of habitats)

Besides decision support, the rationale behind such a holistic assessment methodology is preventing problem shifting, whether it is from one phase of the life cycle to another, from one environmental impact to another, or from one region to another. There is a broad agreement within the scientific community that LCA is one of the most appropriate tools for the evaluation of the environmental burdens associated with biofuels and overall bioenergy production (Petersen, 2008; Cherubini *et al.* 2009; European Commission, 2010), or with products in general (Finnveden *et al.* 2009). However, LCA has also been criticized, among others for the discrepancy between the LCA results obtained from different studies assessing similar systems (e.g. Wardenaar *et al.* 2012; Gentil *et al.* 2010) and for being a rather static methodology, not allowing to capture important dynamic effects such as competition and substitution between different uses for the products involved in the studied system (e.g. Petersen, 2008).

It is the postulate of this PhD work that these critics can be overcome through the application of the so-called consequential LCA methodology (c-LCA), a method representing the convergence of LCA and economic modeling methods (Earles & Halog, 2011). As the application of this methodology is the core of this PhD work, an attempt is made to make a concise description of what it consists of.

2.2 Consequential life cycle assessment

There are two essential differences between consequential and "traditional" LCAs (referred to as attributional LCAs, or a-LCA). One major difference consists of the way they handle the "multifunctionality problem" of product systems. In fact, most product systems will not only produce the product of interest, (e.g. milk), but also a variety of co-products along with it (e.g. meat, leather, calf, manure, meat & bone meal, etc.). Both the c- and a-LCA approaches to multifunctionality reduce the multioutput systems to systems with only single output processes, but in a rather different way. The attributional approach, described among others in the LCA ISO standards itself (ISO, 2006a;b), consists to partition the various flows (emissions, resource extractions, etc.) involved in the LCA system between the studied product and its co-products. One key flaw of such an approach is that each method for allocating, whether it is according to products and co-products mass, dry matter (DM) content, energy/exergy content, sale price, or any other parameter, will yield different results, and as such deliver diverging answers (e.g. Cherubini *et al.* 2011a). The lack of any guidance as to which allocation model is right and how cause and effect relationships are reflected by the allocation model leads to confusion about how to understand and interpret the LCA results. Moreover, this method practically always fails to ensure that both the mass, energy and elemental balances are preserved (Weidema & Schmidt, 2010). The LCA ISO standard somehow recognizes the limit of this approach for dealing with the co-products by stating that "whenever possible, allocation should be avoided" (ISO, 2006a;b). On the other hand, c-LCA proposes to handle multifunctionality through a technique termed "system expansion" in the LCA terminology. In system expansion, the boundary of the LCA system is expanded in order to include the market-induced changes inferred in the studied system by an increased/decreased production of these co-products (Suh *et al.* 2010).

For example, in an LCA modeling different alternatives to provide an increased barley production, an a-LCA would seek to partition the environmental flows between the barley and the co-produced straw, while a c-LCA would instead model the environmental impacts of the barley production system as a whole (including the fate of the straw). If, in the system modeled, more barley (and thus straw) production involve that more straw is incorporated to the soil, this would be modeled accordingly in c-LCA. Similarly, if this extra straw rather means that more straw is burned in small-to-medium biomass combined heat and power (CHP) plant, which in turn involves that less fossil fuels are burned to provide the corresponding heat and power, the LCA system would, under a c-LCA, be expanded accordingly in order to include these effects. The latter example illustrates a case where system expansion allows to take into account not only the affected emission flows from the studied product/service itself, but also those of other systems with which the studied system is coupled (in this example, the energy system). The most significant environmental implications of a new technique or managerial procedure may indeed often be found within such relations to adjoining systems (Hamelin & Wenzel, ***III***). Typically involved adjoining systems resulting from an intervention in bioenergy systems are the energy production system, the fertilizer production system and the crop/feed production system.

The other major difference between c-LCA and a-LCA is actually a logical implication of applying system expansion, and regards the type of data included in the LCA model. While a-LCA uses “average data” (e.g. an average of all electricity sources used in a given national electricity mix), c-LCA includes “marginal data” only, i.e. those from the processes and/or suppliers that are responding to changes in demand by corresponding changes in supply. A procedure for identifying marginal suppliers is described in Weidema *et al.* (1999), updated in Ekvall & Weidema (2004), and further refined in Weidema *et al.* (2009). The main flaw of using average data is that suppliers not affected by the decision supported by the LCA will be included in the model. This can be illustrated through the simplified example of an LCA of a fictive product system using electricity from the Nordic grid. In this fictive example, an a-LCA would include the imported hydro-power from Norway. Yet, hydropower is a constrained resource (at least in the Norwegian case), as it cannot be expanded much further due to natural limitations. Therefore, using it for producing one more unit of the particular studied product system means that less hydropower is available for other product systems that were formerly using this hydropower. As a result of Norwegian hydropower not being able to respond to this increased demand for electricity, more electricity from e.g. coal power plants in Denmark has to be supplied. In this simplified example, coal-based electricity would therefore be the marginal supplier to include in the LCA, as it is the one reacting to the production of one additional unit of the studied product system. This example highlighted physical (or natural) constraints of supply, but such constraints may also be of political/regulatory or economic nature (Ekvall & Weidema, 2004).

Constrained products/resources are typically involved in biosystem LCAs, as it will be demonstrated throughout this PhD work. As illustrated in the Norwegian electricity example, a constrained product/resource, if used for the product/service of interest (e.g. system A), is no longer available for the system in which it was used before (e.g. system B), which triggers market reactions leading to various consequences, among which figure the production of a substitute for that system (system B). The production and handling of this substitute should then be included in the LCA model. This procedure may be seen as a parallel procedure to system expansion (which is applied when dealing with co-products) and is further elaborated in section 2.2.1.

In a nutshell, the purpose of the consequential LCA is to show the environmental consequences of the decision that is supported by the LCA (Wenzel, 1998). In other words, it shall reflect that choosing one alternative over another involve an increasing demand for that alternative. The environmental consequences of this are precisely what the consequential LCA aims to model, and this is done through system expansion and the use of marginal data. Consequential LCA thus strives to include, in the product system, only what is affected by a change in demand for the studied product/service alternative. In essence, it endeavors to model the environmental consequences of a decision through the affected chains of economic causal relationships (Ekvall & Weidema, 2004), rather than solely describing the main material/energy flows from raw material extraction to disposal of a given product. In this context, economic casual relationships are at least as important as physical flows (Ekvall & Weidema, 2004).

It should also be highlighted that consequential LCAs are performed under the assumption that the studied changes in demand can be regarded as being very small compared to the overall production in society (Weidema *et al.* 1999), so the change can be analyzed in isolation under the so-called *ceteris paribus* condition (i.e. all other things remaining equal). Further details on c-LCA modeling can be found in Weidema (2003), Ekvall & Weidema (2004) as well as in Weidema *et al.* (2009), while a review of the different consequential LCAs published up to 2011 is presented in Earles & Halog (2011).

From this point forward, unless otherwise specified, the acronym “LCA” will be used in reference to the consequential LCA methodology.

2.2.1 System boundary

Delimiting the system boundary is a crucial step when performing LCAs. It consists to determine which processes should be included in the LCA model, and which processes should not be included. In consequential LCA modeling of a given alternative, any part of the system not affected by a change in demand for the studied product/service should be excluded of the analysis. For example, an LCA on biodiesel from the fat of dead animals should not include the animal production stage in the LCA system, as the animal production is determined by a market that is independent of the demand for more biodiesel of this type. In other words, farmers will never produce more pigs just because more biodiesel based on animal fat is demanded. This rationale applies for wastes in general: as any system generating waste would, of course, be unaffected by the use of the waste, the processes upstream the generation of these wastes should not be considered in the LCA. This also applies for manure, being a waste of the animal production system.

As above-described, system expansion is, in c-LCA, applied whenever co-products are involved. When the co-products are displacing processes that also have multiple products, a cascade of by-products occurs, which shall be included within the system boundary until a single output process is reached. A classic example to illustrate this is the case of soy meal production, detailed in Dalgaard *et al.* (2008) and summarized as follows: increased soy meal production involves increased soy oil (co-)production, causing a decrease in palm oil production (palm oil being identified as the marginal oil), but consequently also the in co-produced palm fruits (a source of both protein and carbohydrates), inducing, somewhere in the world, an increase in the production of a marginal protein & carbohydrate feed crop. System expansion, however, shall not be seen as an indefinite procedure. The expansion shall indeed be stopped at the point where the consequences are so small (or the uncertainties so large), that any further expansion of the boundaries would yield no significant information for decision support (Ekvall & Weidema, 2004).

It section 2.2, it was mentioned that the system boundary must include the reactions occurring when the system to model involves the use of a constrained resource that would have been used in another life cycle otherwise. In this PhD work, this service no longer provided is referred to as the "lost alternative". To illustrate this, the example of biodiesel based on animal fat can be used again. In this case, let's suppose that the animal fat, if not used for biodiesel, would have otherwise been burned for producing heat and electricity (lost alternative), as in (Hedegaard Jensen *et al.* 2007). Diverting the fat towards biodiesel production thus involves that the heat and power no longer produced by the animal fat must be supplied by the marginal sources of heat and electricity. This induced production of heat and electricity occurring as a result of using the animal fat for biodiesel should thus be included in the LCA.

Examples of the establishment of system boundaries for the LCA case studies modeled within this PhD are presented in Tonini *et al.* (***II***) (Figure 1; Figures S1-S11), Hamelin *et al.* (***IV***) (Figure 1) and Hamelin *et al.* (***V***) (Figures S1-S7).

2.2.2 Reference system

Based on the consequential LCA rationale, the results and conclusions of an LCA shall reflect the consequences of choosing one alternative over another. This involves that LCA is always a comparison, between one, two, or more alternatives and a reference system, i.e. the system prevailing under a status-quo situation (no change). More details about the establishment of a reference system are provided for the specific case of manure management LCAs (Hamelin & Wenzel, ***III***; section 7).

2.3 Marginal suppliers' identification

As earlier stated, guidelines have been developed in order to identify the marginal technologies/processes/suppliers to include in the LCA, i.e. those affected by a change in demand (Weidema *et al.* 1999, 2009; Ekvall & Weidema, 2004). These will not be repeated here, but the main guiding principles are presented.

One important principle in determining the marginal supplier consists to distinguish whether the temporal scope of the LCA is “short-term” or “long-term”, where these are defined in terms of production capacity. State-of-the-art literature on consequential LCA (Ekvall & Weidema, 2004; Weidema *et al.* 2009) describe changes occurring on a short-term perspective to influence the capacity utilization only, and those occurring on a long-term perspective to affect the overall total capacity itself (i.e. installation of new units or decommissioning of old units). In other words, the long-term perspective represents a period long enough to include replacement of capital equipment, and conversely for the short-term (Weidema *et al.* 1999). Schmidt *et al.* (2011), Ekvall & Weidema (2004), as well as Weidema *et al.* (2009) however all argue that long-term effects are the most relevant effects for most LCA models, as LCAs are generally used to support decisions having long-term implications. Weidema *et al.* (2009) further suggest that as a default, it should be assumed that the marginal technology is the most competitive one, which they emphasize as being a forefront modern technology. The rationale is that an increase of demand for a given product will, no matter how small it is, contribute to the accumulated trend in the market volume, and as such influence, sooner or later, capital investments towards the implementation of new modern technologies and old technologies being phased out. However, if a short-term perspective is modeled, or if the studied increase in demand applies for a product in a declining market, Weidema *et al.* (2009) suggest to consider that the marginal technology is the least competitive one, which they pinpoint as being the oldest applied technology (this is then labeled as the short-term marginal). In this case, the justification is that since no new capacity is being installed, the consequence of an increased demand simply consists to postpone the decommissioning of an old plant.

Other key guiding principles in the selection of the marginal technologies include the identification of the market geographical scope (local, national, international), and to determine, as earlier mentioned, if constraints (whether natural, political or market-based) on capacity increase applies for the involved technologies. If the production capacity of a technology is fixed by such constraints, it will thus not be affected by any decisions based on the LCA results and as a result cannot be the marginal technology (Ekvall & Weidema, 2004).

Of course, it can be argued that the choice of a given marginal technology, process or supplier is highly uncertain, especially when the long-term horizon is considered. However, as pointed out by Weidema *et al.* (2009), refraining from attempting to model the future does not appear as a viable solution either, since the raison d’être of LCA is to provide answers for supporting decisions to take place in the future. State-of-the art sensitivity analyses considering various alternatives for the marginal suppliers (i.e. those that are the most likely to affect the LCA outcome) probably represents the best compromise to handle this situation, and this was applied in all LCA case studies involved in this PhD thesis (Hamelin/Tonini et al. ***II, IV, V***).

In this PhD work, in which LCA is applied to assess the use of agricultural biomasses for energy purposes, three marginal suppliers are of particular importance, namely the marginal energy (heat and electricity), marginal fertilizer (for nitrogen, phosphorus, and potassium) and marginal crop. These were in fact involved in all LCA case studies that were carried out, and their identification is further detailed below

2.4 Marginal energy

Energy use, i.e. as electricity and heat, is often a critical issue for LCAs. In the last decade, a lot of publications have been dedicated to the question of the marginal energy data that should be included in LCAs (e.g. Sjödin & Grönkvist, 2004; Curran *et al.* 2005; Moora & Lahtvee, 2009; Mathiesen *et al.* 2009; Fruergaard, 2010; Lund *et al.* 2010; Schmidt *et al.* 2011). There are two main reasons why the type of energy included in an LCA is so important: first, because mostly all processes included in an LCA require energy input and second, because the results for the global warming impact are tightly related to the type of energy selected. In the context of bioenergy LCAs, the outcome of the LCA is of course significantly affected by the identification of the energy (in terms of fuel and producing technology) that is substituted by the bioenergy produced.

In spite of this, there is, to the author's knowledge, no publication available summarizing, in one single document, all important energy-related issues with respect to LCA modeling. It is therefore the aim of this section to do so, as concisely as possible.

The most important energy producing technologies and their related input fuels are summarized in Table 1, categorized in function of the type of energy they produce (electricity and heat, electricity only, or heat only). It should be highlighted that this overview is a mere summary for the purpose of LCA; many of the technologies could in fact be subdivided into many more detailed categories (e.g. on-shore and off-shore wind mills, in the case of the wind turbines).

Table 1. Overview of the most important energy producing technologies and their related input fuels[a]

Heat and electricity		Electricity only		Heat only	
Technology	**Fuel**	**Technology**	**Fuel**	**Technology**	**Fuel**
Steam turbine, back-pressure and extracting	Coal, natural gas, biomass, combustible waste, geothermal brine[c], enriched uranium	Steam turbine, condensing	Coal, natural gas, biomass, combustible waste, geothermal brine, enriched uranium	Boilers	Electricity[d], waste, biomass, natural gas
Gas turbine[b]	Natural gas, light oil, non-fossil methane-based gas[d,e], LPG[f]	Fuel cell	Hydrogen-containing gas (e.g. natural gas, methane, methanol, hydrogen)	Heat pump	Heat source (e.g. ambient air, water, ground) and electricity or heat[d,g] (e.g. steam, hot water, flue gas)
Stirling engine	Biomass, natural gas, oil	Solar photovoltaic	Solar radiation	Geothermal heat pump	As for heat pump, where the heat source is heat from brine[c], from underground reservoirs
Gas engine	Natural gas, non-fossil methane-based gas[d,e]	Wave power	Wave	Solar collector	Solar radiation
		Wind turbine	Wind		

[a] Based on the data from Energinet.dk & DEA (2012).

[b] Can be single-cycle, or combined-cycle (gas turbine combined with steam turbine). The former is typically used for supplying power only during peak demand (emergency units), although heat can be recovered from the exhaust gas.

[c] Saline water

[d] For example biogas, syngas, landfill gas, etc.

[e] This is a "secondary" fuel, in the sense that a "primary" fuel (like coal, natural gas, biomass, etc.) was needed to produce this "secondary" fuel

[f] Liquefied petroleum gas (i.e. propane or butane).

[g] Whether an input of electricity or heat is required depends on the type of heat pump: absorption heat pumps are driven by heat while compressor heat pumps are driven by an electricity input.

In most countries, electricity is currently being produced from either hydropower or large steam turbines on the basis of fossil fuels or nuclear power (Lund, 2010). In Denmark, about half of the annual electricity production is provided by large combined heat and power (CHP) plants (Table 2) relying on steam turbines. Therefore, a few basic principles on how they work must be understood in order to properly model CHP in LCA.

Combined heat and power plants powered by a steam turbine are most commonly known as coal, nuclear, or natural gas plants, depending on the type of fuel they use to generate the steam. In these plants, the steam undergoes a cycle, also referred to as the Rankine cycle, consisting of 4 main stages. First, the water is pumped and compressed to the operating pressure of the boiler (stage 1). The boiler acts as a large heat exchanger where the heat originating from the combustion gases (of whether coal, natural gas, biomass, or other sources) is transferred to the water (stage 2). This produces a so-called superheated steam which is directed to a turbine (or a series of turbines). There, the steam expands between layers of turbine blades (stage 3), mounted on the turbine shaft, and thereby produces electricity by rotating the shaft which is itself connected to an electric generator (Çengel & Boles, 1998). During this process, the temperature and pressure of the steam drop considerably, and the steam is then conveyed to a condenser, where its heat is rejected to a cooling medium such as sea water, the water returning from district heating (ca. 40°C), or the atmosphere, in areas where sea water is not available or its use restricted (stage 4). The condensed water is then pumped back to the boiler and the cycle can be repeated over again. A lower cooling medium temperature at the condensing stage allows a lower pressure for the steam at the outlet of the turbine, which in turns involve that more electricity can be produced (European Commission, 2007).

On the perspective of the Rankine cycle, the main difference between coal, natural gas and even nuclear power plant lies essentially at the boiler stage, i.e. a different boiler technology will be used for each. However, each fuel does involve a different logistic (e.g. transport, storage, pre- and post-treatment), which in turn influences the overall design of the plant. In this sense, it does matter to distinguish if a new steam turbine-based capacity will be fuelled by e.g. coal or natural gas, as the plant cannot be converted so easily from one to another.

Three main types of steam turbines may be distinguished: condensing, back-pressure and extraction turbines (Energinet.dk & DEA, 2012). With condensing turbines, the steam fully expands in the turbine, and thereby transfers all of its energy to work[4] generating electricity, the most valuable form of energy (Çengel & Boles, 1998). As a result, the exhausted steam is already in a partially condensed state, at a pressure well below the atmospheric pressure. No heat is produced with condensing turbines, so these are not part of a CHP plant, but rather a power plant (producing power only).

With back-pressure turbines, a higher exhaust steam pressure is purposely set, as this also involves a higher steam temperature, which can be used as a heat source for industrial processes or for district heating. This "configuration" implies that the steam could not expand fully to generate the maximal amount of electricity; less electricity is thus produced as compared to the condensing turbine, although the overall energy production is much higher. With back-pressure turbines, the amount of electricity and heat generated are directly proportional: if less electricity is produced (e.g. because of a reduced demand), less heat will also be produced.

Extraction turbines, on the other hand, are much more flexible and allow to produce heat and electricity without fixed ratios between the two (so 100% electricity and 0% heat may be produced, or vice-versa). This is typically schematized on a so-called "power-heat diagram", as for example illustrated in Energinet.dk & DEA (2012). This means that with these turbines, the unit can, depending on the demand and price conditions, be whether a power plant, a CHP plant, or a district heating plant (i.e. producing heat only). With extraction turbines, steam can be released at various points of the turbine, instead of being released at the exhaust only. Of course, the earlier the steam is released, the less it can expand, and thus the electricity it can produce.

Modeling CHP also involves to distinguish between the type of plant affected, i.e. whether it is a centralized or decentralized plant. According to the Danish Energy Agency (DEA), the former endeavors to produce a maximum of electricity, while the latter aims to produce mostly heat (DEA, 2009). Moreover, centralized CHP plants are typically located in big cities with relatively important population densities, while decentralized CHP plants are found in smaller town. In Denmark, there were (in 2009) 16 centralized CHP plants and over

[4] In Çengel & Boles (1998), work is defined as: "an energy interaction that is not caused by a temperature difference between a system and its surroundings […]. More specifically, work is the energy transfer associated with a force acting through a distance". It can be expressed in kJ (and 1 kJ = 1 N·m = 1 kPa·m^3).

665 decentralized CHP plants (DEA, 2009). As illustrated in Table 2, the greatest share of both electricity and heat delivered in 2011 in Denmark was from centralized CHP plants. Further, for this same year, nearly half of the primary energy used to produce electricity came from coal, while the primary energy used to produce heat came mostly from biomass (Table 3).

Table 2. Heat and electricity delivered in 2011 in Denmark, per type of plant[a]

	Electricity	Heat
Centralized CHP plants	53%	44%
Decentralized CHP plants	13%	19%
District heating (DH) plants (providing heat only)	-	19%
Portion not produced by CHP or DH plants	34%	18%

[a] From DEA (2012)

Table 3. Primary energy sources used to deliver heat and electricity in Denmark (year 2011), per fuel type[a]

Primary energy sources	Electricity		Heat		Total[c]	
	PJ	%	PJ	%	PJ	%
Coal	115	46%	16	19%	130	39%
Natural gas	43	17%	22	26%	65	20%
Biomass[b]	39	16%	35	42%	74	22%
Wind	35	14%	-	0%	35	11%
Waste, non biodegradable	9	4%	7	8%	16	5%
Oil	4	2%	3	3%	7	2%
Biogas	3	1%	0.7	1%	3	1%
Wave	0.06	0%		0%	0.1	0%
Sun	0.05	0%	0.2	0%	0.3	0%
Geothermal brine	0	0%	0.3	0%	0.3	0%
Heat pump	-	-	0.3	0%	0.3	0%
Total	**248**	**100%**	**84**	**100%**	**331**	**100%**

[a] From DEA (2012). It must be highlighted that the values found in this table illustrate the share of primary energy from each resources needed to provide the Danish heat and electricity mix in 2011. The total values, for heat and electricity, are therefore not representing the amount "delivered" (or consumed) only, but also include the losses.

[b] Straw, wood, biowaste, and bio-oil, the latter applying only for heat production.

[c] This does not corresponds to the total Danish energy consumption (which should also include the oil used in transport), but simply to the total use of heat and electricity, in terms of primary energy. Inconsistencies in the total are due to rounding.

2.4.1 Electricity

The key approach of today's electricity system consists to constantly adjust the supply in order to match the electricity demanded at any time[5]. This allows maintaining a constant frequency on the grid, which is required to ensure the stability of the electricity network and prevent damages to equipment. In Denmark, the task of securing this balance between electricity demand and supply has so far only been managed by large power stations (Lund, 2010).

[5] The concept of flexible demand, where products and systems are designed to specifically use electricity in periods of high electricity production, is however gaining growing interest among energy planners. This concept, commonly referred to as "smart grid", as well as its potential opportunities and drawbacks, is however not tackled within this PhD.

The electricity demand, throughout a day (and a year), is characterized by fluctuations periods, where the demand for some specific periods is much higher than the average demand. These periods of very high electricity demand are termed as peak load. Some plants are thus run to provide this peak load demand only, and these require technologies that can easily and quickly be switched on and off. Gas turbines, which meet these desirable characteristics, are typically used for peak load demands (European Commission, 2007). As opposed to peak load plants, base load plants provide the base load for electricity generation. The electricity network also comprises stand-by plants that are paid to be ready for eventually being used as a backup if necessary (e.g. to compensate for any failure at a given unit). These are referred to as ancillary services.

In the perspective of the earlier described short-term and long-term marginals, the short-term marginal electricity is generally identified as the peak-load technology (Weidema *et al.* 1999; Moora & Lahtvee 2009), being also the technology with the highest variable costs of all running plants (Sjödin & Grönkvist, 2004). Usually, wind power (when blowing), hydropower and nuclear plants are likely to be operated as base-load suppliers when the electricity sale price is low[6], as they allow to produce electricity with relatively low variable costs, while CHP plants will operate as the price increases (Sjödin & Grönkvist, 2004; Moora & Lahtvee, 2009; Lund *et al.* 2010). In fact, as explained in (Lund, 2010), Danish CHP plants have been designed with relatively high production and storage capacities, making it possible to produce mainly during high-tariff period. When electricity sales prices are high, CHP units thus operate at full capacity and store the heat, which can be supplied when prices decrease. In the “merit of order” illustrated in Sjödin & Grönkvist (2004) as well as in Lund *et al.* (2010) for the Nordic electricity, coal condensing would be the technology operated after CHP plants, followed by gas or oil condensing plants, while the more expensive gas turbines would be used last. Further, both studies showed that the annual demand curve cut the supply curve within the coal-condensing segment of the supply.

Several studies with a Danish or Nordic scope similarly identified condensing coal power plants as the marginal electricity source (e.g. Mattsson *et al.* 2003; Weidema, 2003; Møller *et al.* 2008). Weidema (2003) however specified a time span of 10 years (therefore up to 2013) for coal-based technologies to represent the marginal electricity. Fruergaard & Astrup (2011), on the other hand, identified coal from large centralized CHP plants as the marginal electricity while Schmidt *et al.* (2007) considered natural gas. Based on a comprehensive energy system analysis considering the 8760 hours of energy production within a year in Denmark, (Lund *et al.* 2010) developed a mix electricity marginal consisting of 49% coal condensing power plants, 18% natural gas condensing power plants, 16% natural gas small CHP plants, the rest consisting of wind as well as large CHP based on coal and natural gas[7].

According to Schmidt *et al.* (2011), the identification of the marginal electricity in the above-mentioned studies is problematic because they relate to the short-term marginal only. Schmidt *et al.* (2011) claim that the long-term marginal electricity suppliers of a country should rather be defined as the national mix of planned or predicted new installations over a specified period of time.

Although long-term marginal electricity technologies are generally seen as those that should be included in a LCA (Ekvall & Weidema, 2004; Weidema *et al.* 2009; Fruergaard, 2010; Schmidt *et al.* 2011), there are much more discrepancies regarding its identification, as well as much less scientific evidence. Weidema *et al.* (1999) refer to a supplier of base-load electricity as the eventual long-term marginal electricity technology. Fruergaard (2010) claims that coal combustion is a likely long-term marginal fuel, although the arguments supporting this are seldom explained. On the perspective of the Danish government’s energy policy

[6] Nowadays, the electricity sector of the European Union is characterized by an open market, which is the result of various EU Directive fostering liberalization (European Commission 2012). This means that electricity is traded on a free market, which in Denmark is the Nordic electricity market Nord Pool. The Nord Pool spot exchange areas include Denmark, Sweden, Norway, Finland, Estonia and Lithuania (Nord Pool Spot 2011), although the two latter will be fully integrated into Nord Pool in 2015 only. Denmark also trades electricity with Germany.

[7] These values apply for the case where an increase capacity of natural gas condensing plants is considered.

milestones stating that coal shall be phased out by 2030 (and reduced by 65% in 2020 in comparison to "today's" use) (Danish Government 2011), this however appears less realistic. According to Thomson & Booluck (2010), governments are likely, when considering new investments, to prefer to burn natural gas, this being a much less CO_2 intensive fuel than coal (Table 4). In fact, coal consumption in Denmark as well as in the other countries around the Baltic Sea (Baltic Sea Regions) has been on a downward trend since 1985, and these countries all transitioned towards the use of more natural gas, hydropower and/or renewables to generate electricity (Thomson & Booluck, 2010). Moora & Lahtvee (2009) similarly pinpoint natural gas as the long term marginal for the Nordic region, also in the perspective of lowering the emission levels. They further forecasted a long-term electricity marginal for the Baltic States (Lithuania, Latvia and Estonia) consisting of natural gas and coal fired power plants (current trend scenario) or of biomass and wind power (sustainable scenario).

In the light of the renewable energy ambitions of Denmark (100% renewable energy for electricity and heat by 2035, and for the whole energy system by 2050; wind to represent 52% of the electricity consumption by 2020) (Danish Government, 2011), and considering that neighboring countries also have long-term targets for fossil free electricity (e.g. Germany, Sweden) (Klaus *et al.* 2010; World Nuclear Association, 2012b), a long-term fossil free marginal electricity can be envisioned as realistic. Wind power is likely to have a high share in such marginal, but being intermittent, could not react alone to a change in demand. As in the situation described by Moora & Lahtvee (2009), a mix marginal consisting of wind and biomass could therefore appear as realistic. Lund *et al.* (2011) actually describe the electricity to consist of 47% wind and 53% CHP (which itself is essentially fuelled by biomass), in a renewable energy future in Denmark. Habib *et al.* (*submitted*), in a study applying for waste management in Aalborg, similarly considered a marginal electricity consisting of 49% wind and 51% biomass. In the case of such a wind-biomass marginal electricity, the marginal biomass would likely consist of wood chips and/or perennial energy crops (as manure, straw, wood pellets are "secondary products" emerging from other activities, and as such would fail to react to a demand increase, and thereby to be the biomass marginal).

Hydropower and nuclear power have not been discussed as eventual long-term (or short-term) marginal. As earlier mentioned, hydropower is limited by the areas available for establishing new plants, and because of this cannot be the marginal electricity technology due to its restrained capacity to respond to a demand increase (Moora & Lahtvee, 2009; Weidema *et al.* 2009). For this reason, it was not included in the above-discussion. Similarly, establishing a new nuclear capacity is not a straight-forward process, and there can easily be a lag of 10 years or more between the decision to establish the new capacity and the moment where it is fully operational (Moora & Lahtvee, 2009). Moreover, it appears unlikely that more nuclear capacities will be installed in reaction to an increased demand of electricity in Denmark. The nuclear electricity in the Nord Pool comes (and will come) essentially from Sweden, Finland, and Lithuania. Sweden, although it repealed its decision taken in the 80's to phase out nuclear power, will only replace its existing 10 operating reactors (World Nuclear Association 2012a), nuclear electricity being on this sense constrained. Similarly, Lithuania phased out its two nuclear reactors, and plans to build a new one to be operational by 2020, while Finland has 4 reactors, one under construction, and two under a planning phase (World Nuclear Association 2012b, 2012c). In other words, investments in additional nuclear capacities are, in the Nordic grid, likely to emerge from Finland only, and on this sense nuclear is rather limited.

In a nutshell, modeling the electricity substituted by the bioenergy produced, although it may at first appear trivial, is not straightforward, especially if CHP units are involved, as further discussed in section 2.4.2 (heat). However, based on Table 4, it is very clear how the choice of the marginal electricity will influence the global warming results: displacing lignite, oil shale or coal will result in greater benefits from bioenergy production, while displacing natural gas or wind would result in smaller benefits.

In this PhD thesis, different marginal have been considered in the LCA case studies performed: an adapted version of the mix marginal developed by Lund *et al.* (2010) (Hamelin *et al.* ***IV***) and coal condensing power plants (Hamelin/Tonini *et al.* ***II, V***). However, a sensitivity analysis with natural gas condensing plants was considered in Hamelin/Tonini *et al.* (***II, V***), which illustrated the magnitude this choice had on the global warming LCA results. These, however, are all short-term marginals. The long term perspective (e.g. a biomass marginal) has thus not been considered, and this is further discussed in section 10.4.5.

In this project, the environmental impacts due to the (avoided) production of electricity were modeled using the following Ecoinvent processes (all described in Dones *et al.* 2007):

i. Coal electricity from condensing power plants: Electricity, hard coal, at power plant/NORDEL U[8] (where 8.66 MJ of coal is burned for producing 1 kWh of electricity): Hamelin/Tonini *et al.* (***II, IV, V***).
ii. Natural gas electricity from condensing power plants: Electricity, natural gas, at power plant/NORDEL U (where 8.68 MJ of natural gas is needed for producing 1 kWh electricity): Hamelin/Tonini *et al.* (***II, IV***).
iii. Wind-based electricity: electricity, at wind power plant/RER U: Hamelin *et al.* (***IV***) (represented 1% in the mix marginal derived from the energy system analysis of Lund *et al.* 2010)

Table 4. Global warming potential of electricity production, per fuel type, not including the transmission network. Only the countries with which Denmark interacts were considered.

Provision of electricity	**kg CO_2 eq. per kWh electricity**	**Comment**
Lignite	1.22	Ecoinvent database[a]. Data for Germany, year 2000.
Oil shale	1.0 – 1.6	EASAC (2007). Minimum data for future plants, maximum data for current plants. Data for Estonia.
Hard coal	0.96 - 1.08	Ecoinvent database[a]. Data for year 2000. Minimum data for NORDEL[b], maximum for Germany.
Oil	0.52 - 1.13	Ecoinvent database[a]. Minimum data for Finland, maximum for Germany.
Natural gas	0.56 – 0.60	Ecoinvent database[a]. Data for year 2000. Minimum data for Germany, maximum data for NORDEL[b].
Wind	0.0113 – 0.0144	Ecoinvent database[a]. Minimum data for on-shore (average European, 800 kW), and maximum for off-shore (2MW, based on Danish wind park Middelgrunden).
Nuclear	0.0095 - 0.0104	Ecoinvent database[a]. Data for year 2000, for Germany. Minimum data for pressure water reactor, maximum for boiling water reactor.
Hydropower	0.0049 – 0.0264	Ecoinvent database[a]. Minimum data for Germany, maximum for Finland.

[a] The Ecoinvent database is described in Frischknecht & Rebitzer (2005), while the electricity data of the Ecoinvent database are documented in Dones *et al.* (2007).
[b] Nordic Countries Power Association.

2.4.2 Heat

As opposed to electricity, heat is traded on a local market. Depending on the specific area where bioenergy-based heat is produced, the fuel substituted may be coal or natural gas from centralized or decentralized CHP plants, or it may substitute any fuel burned in an individual boiler if the heat is produced in an area where district heating is not available.

[8] This is the process used to model the avoided energy production from power plants. When modeling the electricity input to processes, the transmission network was also accounted for. For this, the Ecoinvent process “electricity, high voltage, production NORDEL, at grid/NORDEL U” was used, and modified by replacing the electricity input by the one selected here (this applies for both the coal and natural gas case). Medium and low voltage electricity processes were created with the same procedure as for high voltage. Low voltage electricity is the one used to model the electricity input to households and service industry (while high and medium voltage were here only needed as intermediaries for creating the low voltage electricity).

In Denmark, a wide-spread district heating network that supplies ca. 60% of the households exists (Dansk Fjernvarme, 2012). With such infrastructure where the heat is produced centrally and distributed to each household, efficiencies much greater than what would be observed if each household would have its own boiler are obtained. Further, district heating allows to use energy that would otherwise be wasted, thereby increasing the efficiency of CHP plants from around 40% (condensing plants) to around 90% (back-pressure or extracting plants). Of course, one pre-requisite for this to hold true is that there is a demand for the heat produced, which is not necessarily the case in e.g. South European countries, or even in Denmark in the summer months.

Most of the heat consumed in Denmark nowadays is produced through centralized CHP plants (Table 2), which, as earlier mentioned, endeavor to maximize their electricity production rather than their heat supply. On this basis, the heat produced from centralized CHP plants could not be the marginal as the process is not run for producing heat (heat is here a co-product).

On a short-term perspective, the marginal heat will, based on the principles described in section 2.3, be supplied by the unit the most likely to be phase out next. This, based on figures from the Danish Energy Agency (DEA, 2011), could likely be a fuel oil or natural gas fuelled district heating plant, given the much higher prices for these fuels, today and in the future, as compared to other fuels used for heat production (coal being by far the cheapest, followed by straw, wood chips and willow). A district heating plant is pinpointed here rather than a decentralized CHP plant, as the CHP unit is likely to be more profitable (i.e. competitive), given the electricity it also produces.

In this PhD work, the marginal heat was, as for electricity, considered in a short-term perspective. In the three case studies performed (Hamelin/Tonini *et al.* ***II, IV, V***), both coal and natural gas district heating plants have been considered as the marginal heat producing technology, whether for the baseline scenarios or as a sensitivity analysis.

On a long-term perspective, considering that both fuel oil and coal are to be phased out in Denmark by 2030 (Danish Government, 2011), and considering the forecasted price of fuels (DEA, 2011), it appears likely that biomass (wood chips or willow, as non-constrained) could be the long-term marginal for heat. If this is from a decentralized CHP plant, then the system studied must be expanded in order to account for the effects that the changes in heat produced have on electricity. To this end, it must be identified if the decentralized CHP plant operates a back-pressure or extracting steam turbine. If a district heating plant producing heat only, or an individual boiler is affected, the modeling is then more straight-forward.

In this project, the environmental impacts due to the (avoided) production of heat were modeled using the following Ecoinvent processes (all described in Dones *et al.* 2007):

i. Natural gas district heating:
   - Heat, natural has, at boiler condensing modulating > 100 kW/RER U (where 0.98 MJ of natural gas is burned for producing 1 MJ of heat): Hamelin *et al.* (***IV***).
   - Heat, natural gas, at boiler atmospheric low-NOx non-modulating < 100kW/RER U (where 1.09 MJ of natural gas is burned for producing 1 MJ of heat): Hamelin/Tonini *et al.* (***II, V***).

ii. Coal district heating:
   - Heat, at hard coal industrial furnace 1-10MW/RER U (where 1.25 MJ is burned to produce 1 MJ heat): Hamelin/Tonini *et al.* (***II, V***).

2.5 Marginal NPK fertilizers

Fertilizers are an input in all LCA inventories and case studies performed within this PhD work (Tonini *et al.* ***II***; Hamelin *et al.* ***I***, ***IV, V***), and for this reason, they were carefully selected in order to include the marginal fertilizers only. The three primary plant nutrients, i.e. nitrogen (N), phosphorus (P) and potassium (K), which are generally the most needed by plants, were considered. Although commonly used as a fertilizer, manure is not considered in the identification of marginal N, P and K fertilizers, as, being a waste product, its production is not influenced by changes in demand for fertilizers.

2.5.1 Nitrogen fertilizer

A variety of mineral N fertilizers are available, some providing N independently of other primary nutrients, and some providing both N and P (e.g. ammonium nitrate phosphate), N and K (e.g. potassium nitrate), or N, P and K (so-called NPK fertilizers). Compound PK fertilizers can also be found. However, based on EFMA (2004), 78 % of the mineral nitrogen is applied through “straight” fertilizers (providing N without P and K) in Western Europe countries, the remaining being applied in multi-nutrient compound fertilizers. For the developing countries, compound fertilizers represent only 14% of the mineral N fertilizers applied (EFMA, 2004). Therefore, straight N fertilizers are used in this study. This also simplifies the inventory building. In Table 5, an overview of the various straight N fertilizers available is presented.

Table 5. Overview of available straight N fertilizers[a]

Fertilizers derived from ammonia and nitric acid	Fertilizers derived from ammonia	Others

Ammonium nitrate (AN):
• Synthesized through acid-base reaction of nitric acid and ammonia in aqueous solution.
• Mixing an AN solution with a sulphate source, calcium source or magnesium source can yield the following fertilizers:
  • **Ammonium sulphate nitrate (ASN)**
  • **Calcium ammonium nitrate (CAN) and calcium nitrate**
  • **Magnesium ammonium nitrate (MAN)**
	Urea:
	• Ammonia and CO_2 are reacted under high pressure (1.2 to 2.8 MPa) and high temperature (175 to 210 °C). The partial dehydration of ammonium carbamate formed in the reactor produces urea as well as water.
	
	Ammonium sulphate:
	• Produced mostly through reaction between ammonia and sulphuric acid.
		Sodium nitrate:
		• Natural sources from Chilean deposit (caliche)[b]
		
		Calcium cyanamide (CaN_2):
		• Made from limestone ($CaCO_3$) and coke.

Urea ammonium nitrate (UAN): • Solution of UAN are produced from urea and AN	**Ammonium chloride** • Typically synthetized through the “double-salt process”, where a sodium chloride solution is reacted with CO_2 and ammonia.	

[a] This Table was made based on Longacre *et al.* (2010). It does not pretend to cover all straight N fertilizers available, but aimed to at least capture the most used ones.
[b] May also be produced synthetically, involving nitric acid or ammonia, according to the synthesis process used.

On the basis of discontinued statistics from FAO (FAOSTAT, 2012), nitrate & ammonia based straight N fertilizers represented ca. 15% of the world consumption in 2002 (the latest year of data), while this was 85% for ammonia only-based fertilizers (and 0.02% for sodium nitrate and calcium cyamide). Based on this, it appears clear that two main types of N fertilizers may be distinguished: those based on ammonia and nitric acid, and those based on ammonia only. Sodium nitrate and calcium cyamide are thus disregarded as potential marginal N fertilizers.

In Europe (EU 25 plus Norway and Switzerland), about 44% of the consumed N fertilizers are nitrate-based (AN and CAN) (EFMA, 2004). The statistics from the international fertilizers association (IFA) show similar figures for Western Europe, where AN and CAN represented 48% of the straight N fertilizers consumed in 2010, closely followed by urea (45%) (IFA, 2012). For the world, statistics from IFA (2012) indicate that urea represented 75% of the straight N fertilizers consumed in 2010. In Denmark, CAN represented over 60% of all N fertilizers consumed in 2009 (including compound fertilizers) (Nielsen *et al.,* 2011). One reason why nitrate-based fertilizers are more used in Europe as compared to the rest of the world relates to the efforts towards ammonia emission reductions undertaken within the EU[9]. Moreover, this is also because nitrate-based fertilizers are readily absorbed by plants and thereby, are more suitable than urea for the cold European springs, the latter needing to be first transformed into plant available forms (ammonium and nitrate) through biological processes (favored among other by warmer temperatures). Under Southern European conditions, nitrate-based fertilizers are also the dominant form in most areas, as many South European regions practice fertigation (adding fertilizers to irrigation water), for which the highly soluble nitrate-based fertilizers are much more suitable than urea (personal communication with Christian Pallière, Director of Agriculture and Environment, European Fertilizer Manufacturers Association[10], October 1st, 2010).

Based on this overview of N fertilizers consumption in Europe and in the world, two N fertilizers may be distinguished as the potential marginals, depending on the market considered: urea (global market) and nitrate-based fertilizers (CAN or AN) (European market). In this PhD work, both fertilizer types were considered. In Hamelin *et al.* (***IV***), AN was considered as the marginal N fertilizer, while CAN was considered in Hamelin/Tonini *et al.* (***I, II, V***). In Hamelin *et al.* (***I***), a sensitivity analysis with urea was made.

In order to determine if “new” or “old” technology for producing these should be considered, the market trend (i.e. increasing or decreasing), should be identified (Weidema *et al.* 2009). Consumption of nitrogen mineral fertilizer has, in Denmark as well as in the EU, experienced an overall significant decrease since 1989 (EFMA 2009; FAOSTAT 2012a; Statistics Denmark 2012a). This is, among others, the result of enhanced environmental regulations. Recent forecasts towards 2020 and 2030 however indicate increases of mineral N use for both the world and in Europe as summarized in Table 6.

Table 6. Summary of available projections for mineral N consumption in the EU and the world

Projection	**Time Horizon**	**Scope**	**Reference**
Mineral N consumption of 14.9 M tonnes[a]	2015	EU	Tenkorang & Lowenberg-Deboer (2009)
Mineral N consumption of 15.3 M tonnes[a]	2030	EU	Tenkorang & Lowenberg-Deboer (2009)
Annual increase in mineral N demand of 1.3 %	2015	EU	FAO (2011)
Increase in N consumption from 10.5 M tonnes in 2011/2012[b] to 10.8 M tonnes in 2021/2022 (annual increase of 0.34%)	2020	EU	Fertilizers Europe (2012b)
Mineral N consumption of 115.4 M tonnes[c]	2015	World	Tenkorang & Lowenberg-Deboer (2009)
Mineral N consumption of 137.4 M tonnes[c]	2030	World	Tenkorang & Lowenberg-Deboer (2009)
Mineral N demand of 112.9 M tonnes	2015	World	FAO (2011)

[a] From a level of 10.4 M tonnes in 2005.
[b] The reference period actually cover the whole period from 2009/2010 to 2011/2012.
[c] From a level of 90.7 M tonnes in 2005.

In the light of the studies summarized in Table 6, it is concluded that N fertilizers is not a declining market, which involves, based on the consequential LCA principles presented in section 2.3, that the most

[9] Urea has a much higher NH_3 emission factor (0.13 kg NH_3-N per kg N applied) than calcium ammonium nitrate (0.01 kg NH_3-N per kg N applied), based on data from Nielsen *et al.* (2009). In fact, two moles of ammonia have the potential to be formed per mole of urea, in the presence of urease enzyme (Mobley & Hausinger, 1989). As opposed to nitrate-based fertilizers, urea cannot be directly absorbed by the plants and must first be hydrolyzed to ammonium by soil enzymes, which involves a pH increase, and thereby considerable losses of N as ammonia (Fertilizers Europe 2012a).

[10] The European Fertilizer Manufacturers Association (EFMA) is now called Fertilizers Europe.

competitive supplier is the one affected by a change of demand[11]. As a result, it is considered that ammonia, the key input for both CAN and urea production, is produced through the so-called steam reforming process[12] (using natural gas), which is much more energy efficient than the partial oxidation process (using heavy hydrocarbons and/or coal) (European Commission 2007).

The data for both urea and calcium ammonium nitrate production are taken from the Ecoinvent processes “Calcium ammonium nitrate, as N, at regional storehouse/RER U” and “Urea, as N, at regional storehouse/RER U”, described in Nemecek & Kägi (2007). These represent European data. However, the marginal producing technology for urea is not likely to be European. In fact, EFMA (2004) specifies that new capacities (for ammonia, from which 97 % of the N fertilizers are derived) are developed in high-demand developing countries (South Asia and China), where the social and environmental legislation is less constraining than in other countries and where cheap supply of natural gas is available. However, no data are available for these, so European data were used as a best proxy.

As presented in Table 5, the production of CAN requires nitric acid, and this is included in the above-mentioned Ecoinvent process. However, the data for nitric acid production in the Ecoinvent database are from 1997, and as a consequence do not represent state-of-the-art technology. For example, Börjesson & Tufvesson (2011) mention that approximately half of the nitrogen fertilizer plants in Western Europe have installed catalytic cleaning equipment (reducing N_2O by some 80 %), and they further add that within the next few years, all plants are expected to have such technology. The authors report an average emission of 3 g N_2O per kg N produced for these plants. Using, based on the Ecoinvent data, an amount of 2.25 kg nitric acid per kg of calcium ammonium nitrate (as N) produced, this represents an amount of 0.0013 kg N_2O per kg nitric acid produced. The actual Ecoinvent process uses 0.00839 kg N_2O per kg nitric acid, which is about 6 times higher. The EU BREF[13] document on the manufacture of large volume inorganic chemicals (European Commission 2007) indicates emission values between these 2 extremes, for plants across Europe. In fact, the data for 42 plants (European Commission 2007)[14] presented in the BREF lead to an average of 0.0062 kg N_2O per kg nitric acid. The best available techniques (BAT) level stated in the BREF is 0.00012-0.00185 kg N_2O per kg nitric acid for existing plants[15]. For new plants, the upper limit is 0.0006 kg N_2O per kg nitric acid. However, FAO (2011) indicates that no increase in ammonia capacity is expected in Western and Central Europe between 2011 and 2015, which consequently may indicate little development in terms of new nitric acid plants in Europe. As a result, these BAT levels new plants were not considered.

In this study, an emission value of 0.0062 kg N_2O per kg nitric acid has been used. This may be seen as a higher end-of-interval value as it is well above the BAT emission levels for existing plants, but it is still about 26 % lower than the original value displayed by the Ecoinvent database. It is judged to represent what could be expected for the new capacities to be built.

Ammonia, the substance at the core of nearly all mineral N fertilizers consumed, is today produced synthetically through the so-called “Haber-Bosch process”, a high pressure catalytic process using, as an N source, the N from air. In Europe, natural gas is generally the most competitive source for the needed hydrogen (H) (EFMA, 2009), so the hydrogen is produced through the steam reforming process, as earlier mentioned. 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

[11] It is however acknowledged that, for N fertilizers, the trend may not only be market driven, but also influenced by a number of factors like agro-environmental measures or other policy intervention types (e.g. Common Agricultural Policy – CAP - measures in EU countries), which are common in the agricultural sector.
[12] Depending on the type of fossil fuel, two different methods are mainly applied to produce the hydrogen needed for ammonia production: steam reforming or partial oxidation. As explained next page, ammonia itself is actually produced through the Haber-Bosch process.
[13] Reference document on Best Available Technique produced under the Industrial Emission Directive (EC, 2010).
[14] Table 3.7.
[15] With a split view stating that the upper limit should be 0.0025.

represents between 50 to 70 % of the total feedstock cost, meaning that the cost of natural gas is an important parameter in the price of N fertilizers. In the light of this, it appears that the ultimate product affected by a change of demand in N fertilizers (under a rising trend) is the supply for H to synthesize the ammonia necessary to produce all fertilizers. This creates a direct interaction with the energy sector, as the gas that is used for the fertilizing industry is not available for competing energy uses. This could in turn trigger a panoply of different reactions whose forecast and/or analysis is beyond the scope of this PhD work, although certainly worth assessing (e.g. fostering shale gas development, fostering investments into water electrolysis technologies, fostering the development of bio-hydrogen technologies, etc.).

2.5.2 Phosphorus fertilizer (P_2O_5)

Within the fertilizer industry, P fertilizers are typically expressed in terms of phosphorus pentoxide (P_2O_5), although P_2O_5 itself is not used as a P fertilizer. This nomenclature will also be used in the present section. Like in the case of N, forecasts for P demand also tend towards an increasing trend, both at the EU and world level, as summarized in Table 7.

Table 7. Summary of available projections for mineral P consumption in the EU and the world

Projection	Time Horizon	Scope	Reference
Mineral P consumption of 4.3 M tonnes[a]	2015	EU	Tenkorang & Lowenberg-Deboer (2009)
Mineral P consumption of 5.2 M tonnes[a]	2030	EU	Tenkorang & Lowenberg-Deboer (2009)
Annual increase in mineral P demand of 1.7 %	2015	EU	FAO (2011)
Increase in P_2O_5 consumption from 2.4 M tonnes in 2011/2012[b] to 2.6 M tonnes in 2021/2022 (annual increase of 0.9%)	2020	EU	Fertilizers Europe (2012b)
Mineral P consumption of 43.8 M tonnes[d]	2015	World	Tenkorang & Lowenberg-Deboer (2009)
Mineral P consumption of 52.9 M tonnes[d]	2030	World	Tenkorang & Lowenberg-Deboer (2009)
Annual increase in mineral P demand of 1.9 %	2015	World[c]	FAO (2011)
Mineral P consumption growth rate of 2.5% annually	2017	World	USGS (2012)

[a] From a level of 3.1 M tonnes in 2005.
[b] The reference period actually cover the whole period from 2009/2010 to 2011/2012.
[c] FAO (2011) however specifies that the global increase comes essentially from Asia.
[d] From a level of 36.6 M tonnes in 2005.

In the light of the studies summarized in Table 7, it is concluded that P fertilizers are, like N, not a declining market, so the “modern” technologies should be considered. As a result, the so-called “wet process[16]” is considered for the production of phosphoric acid (which is at the core of most P fertilizers), rather than the “electric furnace process”, which is much more energy intensive and which would represent, in the case of a decreasing trend, the “least competitive unit”. As shown in Table 8, there are 2 main types of mineral P fertilizers: based on phosphate rock directly, or based on phosphoric acid (itself made from phosphate rock).

[16] Practically all the phosphoric acid used in modern fertilizer production nowadays is made by the wet process. In this process, the phosphate rock is acidulated with strong mineral acids, the most commonly used being sulfuric acid, and this reaction produces phosphoric acid along with calcium sulfate (Longacre *et al.* 2010).

Table 8. Overview of available P fertilizers[a]

Fertilizers derived from phosphoric acid	Fertilizers derived from phosphate rock directly
Triple superphosphate (TSP): • Produced from acidulation of ground phosphate rock with phosphoric acid. Its typical P_2O_5 concentration is 40-49%. Its chemical formula is $H_2PO_4^{-1}$.	**Superphosphate (or single superphosphate):** • Produced through a reaction involving concentrated sulphuric acid, water and phosphate ground rock. Its typical P_2O_5 concentration is 16-22%. Its chemical formula is $Ca(H_2PO_4)_2 \cdot H_2O$.
Mono- and di- ammonium phosphate (MAP and DAP): • Produced from the reaction of anhydrous ammonia and phosphoric acid. The chemical formula is $NH_4H_2PO_4$ for MAP and $(NH_4)_2HPO_4$ for DAP. MAP can also be produced by reacting (single) superphosphate and ammonia.	

[a] This Table was made based on Longacre *et al.* (2010). It does not pretend to cover all P fertilizers available, but aimed to at least capture the most used ones.

According to Longacre *et al.* (2010), superphosphate represented half the world production of P fertilizers up to 1962, but is seldom used today, due to its low P_2O_5 concentration and to the rising cost of freight. Based on statistics from the International Fertilizer Association, diammonium phosphate was, in both "Western Europe" and the world, the P fertilizer with the greatest apparent consumption for the period 1999-2010 (IFA, 2012) (compared to monoammonium phosphate and triple superphosphate). FAO (2009) reports that close to 40 new monoammonium phosphate, diammonium phosphate and triple superphosphate units shall be constructed in the short-term future in 10 different countries, and that nearly half of it should be in China. Other facilities are also planned in Africa, West Asia, East Asia and Latin America. FAO (2009) also specifies that most of these 40 new units should be diammonium phosphate units. Based on this, diammonium phosphate (DAP) was considered to be the marginal P fertilizer in this study[17].

The life cycle inventory data for DAP production were taken from the Ecoinvent database[18] Nemecek & Kägi (2007). In this database, two main processes are available for the production of the phosphoric acid, distinguishing whether the phosphate rock is mined from Morocco or the US. The background Ecoinvent report for phosphoric acid (Althaus *et al.* 2007) indeed highlights two main types of refining processes (so-called beneficiation) for the mined phosphate rock: as done in the US (wet beneficiation[19], resulting in 29 % P_2O_5 in the beneficiated rock) and as done in Morocco (dry beneficiation, resulting in 33 % P_2O_5 in the beneficiated rock). In this study, the phosphate rock mining (and beneficiation) process used is the one from Morocco, since it is the place where the largest economically extractable reserves are located (USGS, 2012), and since phosphoric acid plants are typically located in the vicinity of phosphate mines (Althaus *et al.* 2007).

Though DAP is primarily a P fertilizer, it also provides N. This must be considered when accounting for the fertilization. In this study, this was accounted for as follows. First, the N and P requirements of the crops were established (Hamelin *et al.* ***I***). Second, the P needs were balanced, considering that there is 0.46 kg P_2O_5 per kg DAP (Nemecek & Kägi, 2007)[20]. From this, the amount of N simultaneously applied was known, based on a content of 0.18 kg N per kg DAP (Nemecek & Kägi, 2007). This amount could then be deducted from the total N to be applied.

[17] In Hamelin *et al.* (***IV***), however, triple superphosphate was used, in order to use a straight fertilizer for simplifying the assessment. The rationale was that triple superphosphate was identified as the most used straight P fertilizer in the EU, based on Schmidt (2007).
[18] Process "diammonium phosphate, as P_2O_5, at regional storehouse/RER U".
[19] It is the beneficiated rock that is used for producing phosphoric acid, and the chemical composition of the beneficiated ground rock will vary according to the beneficiation process applied. Not to be confounded with the "wet process" for producing phosphoric acid.
[20] First, the amount of P needed had to be expressed as P_2O_5, and this was done as follows: amount of P needed, as P_2O_5= amount of P needed, as P × (molecular weight of P_2O_5 / 2 × molecular weight of P), where the molecular weight of P_2O_5 is 141.943 and the molecular weight of P is 30.974. Then, the amount of DAP applied could be calculated: DAP applied = amount of P needed, as P_2O_5 / 0.46 kg P_2O_5 per kg DAP.

2.5.3 Potassium fertilizer (K_2O)

Within the fertilizer industry, K fertilizers are typically expressed in terms of potassium oxide (K_2O), although K_2O itself is not used as a K fertilizer. This nomenclature will also be used in this section.

As for N and P fertilizers, available projections indicate an increasing trend for K fertilizers consumption, both in Europe and worldwide Table 9.

Table 9. Summary of available projections for mineral K consumption in the EU and the world

Projection	Time Horizon	Scope	Reference
Mineral K consumption of 5.0 M tonnes[a]	2015	EU	Tenkorang & Lowenberg-Deboer (2009)
Mineral K consumption of 6.0 M tonnes[a]	2030	EU	Tenkorang & Lowenberg-Deboer (2009)
Annual increase in mineral K demand of 3 %	2015	EU	FAO (2011)
Increased in K_2O consumption from 2.7 M tonnes in 2011/2012[b] to 3.2 M tonnes in 2021/2022 (annual increase of 1.8%)	2020	EU	Fertilizers Europe (2012b)
Mineral K consumption of 28.5 M tonnes[c]	2015	World	Tenkorang & Lowenberg-Deboer (2009)
Mineral K consumption of 32.8 M tonnes[c]	2030	World	Tenkorang & Lowenberg-Deboer (2009)
Annual increase in mineral K demand of 3.1%, from 2011	2015	World	FAO (2011)
Mineral K consumption growth rate of 4% annually	2017	World	USGS (2012)

[a] From a level of 3.2 M tonnes in 2005.
[b] The reference period actually cover the whole period from 2009/2010 to 2011/2012.
[c] From a level of 26.6 M tonnes in 2005.

Table 9' projections highlight that as for N and P mineral fertilizers, the market for K fertilizers is not in a declining trend. Yet, in 2009, the demand for potash dropped to its lowest level since the past 30 years (Heffer & Prud'homme, 2009). However, according to FAO (2011), strong demand prospects in the medium-term have prompted many prospective producers to invest in potash projects worldwide. This new capacity is forecasted to be entirely in the form of muriate of potash (MOP), i.e. potassium chloride (KCl).

Varieties of potassium fertilizers include potassium chloride, potassium sulphate and potassium nitrate. However, potassium chloride accounts for about 95 % of all potassium fertilizers used in agriculture, being the cheapest per tonne Johnston (2003)[21]. Potassium chloride is therefore considered as the marginal K fertilizer in this study.

As opposed to N and P fertilizers, KCl is produced directly from the potash salts, without the need to involve intermediate products, although additional processing is of course involved. Mined raw potash salts are concentrated in order to produce KCl. There are three main processes used to carry out this concentration: thermal dissolution, flotation and electrostatic separation (Nemecek & Kägi, 2007). The choice of the method is function of the raw salt as well as of the final product requirements, and sometimes consists of a mix of different technologies (Kali, 2010). However, according to Johnston (2003), the most commonly used method for carrying out this separation is thermal dissolution.

In this PhD work, the environmental impacts due to the production of KCl were modeled using the Ecoinvent process "Potassium chloride, as K_2O, at regional storehouse/RER U", described in Nemecek & Kägi (2007). This is mainly based on data from Germany, which is the largest supplier of potassium fertilizer in Western Europe (Nemecek & Kägi, 2007). The process includes a mix of the three main used technologies for the concentration of the salt.

[21] Potassium sulphate and potassium nitrate are supplied to crops that are very sensitive to chloride, although the latter, a high-quality and pricy fertilizer, is used with high value crops only (Longacre *et al.* 2010).

2.6 Marginal crop

This PhD investigates the consequences of using agricultural biomass as part of the Danish energy strategy towards 100% renewable energy systems. One key part of this, as earlier mentioned, is energy crops, i.e. crops grown especially for energy purpose. Although these may be imported, this PhD work only assessed the environmental consequences of energy crops being grown in Denmark.

Producing bioenergy from energy crops involves the use of Danish agricultural land in order to grow the crops. In a country like Denmark, where 68% of the total land is used for cropland and where policies have been adopted in order to double the forested area (nowadays representing ca. 13% of the total land) (Nielsen *et al.* 2011), very limited conversion from forest or alike nature types is occurring. Most likely, the land needed to grow the energy crops will be taken from actual Danish cropland, involving that one crop cultivated today will be displaced. Such a displaced crop is, in consequential LCA, referred to as the marginal crop.

Based on findings from Weidema (2003), spring barley is often designated as the marginal crop for Western European countries, being one of the crops with the lowest gross returns (Dalgaard *et al.* 2008; Schmidt 2008; De Vries *et al.* 2012). Further, spring barley is also a rather low-yielding crop, compared to other cereal and coarse grain crops grown in Denmark (Hamelin *et al.* ***I***). In Denmark, the statistics from 1982 to 2011 actually show that the area cultivated in spring barley underwent a drastic decreasing trend as compared to any other crops cultivated on Danish land (Statistics Denmark, 2012), confirming the likelihood of it being the crop displaced by new crops.

Spring barley was thus considered to be the marginal crop in this PhD work (Hamelin/Tonini *et al.* ***II, IV, V***). However, it can be argued that this is a short-term marginal only, and that it does not reflect a high bioenergy future in which the demand for biomass and arable land will likely increase to a large extent. In such a future, it is seen as plausible that the benefits of the greater (and potentially increased) yield of maize, as compared to barley and to most other crops cultivated in Denmark, could change the cropping towards maize for both animal feed and energy. In this future, barley would already be completely offset by maize, so a new energy crop, say *Miscanthus*, would displace maize, the only remaining crop to be displaced. Such long-term marginal crop was used in Hamelin *et al.* ***V*** (where barley was considered in sensitivity analysis).

The life cycle inventory data used for the displaced barley and maize were those presented in Hamelin *et al.* (***I***), which are further detailed in section 6.

2.7 Price elasticity

It is a common practice in consequential LCA to consider, when substitution occurs, that the substitution ratio is 1:1, i.e. that if demand increases with 1 unit, the producers will react by increasing their supply with 1 unit, and conversely when the demand decreases (Weidema *et al.* 2009). This assumption, however, may be seen as over-simplistic. In fact, a change in demand and/or supply will, for most decisions to be modeled by LCA, likely influence prices, which in turn may induce a substitution ratio lower than 1:1.

The responsiveness, or sensitivity, of the quantity supplied of a given good to a change in its price can be quantified in terms of price elasticity. In other words, the elasticity of supply represents the relative change in production per relative change in price. A 1:1 substitution ratio involves a perfectly elastic supply (elasticity of 1), while an inelastic supply would be characterized by an elasticity value of zero (i.e. the supply is not at all affected by changes in prices). Within the economical science, it is attempted, through the use of time series and econometric models, to derive elasticities for a range of specific goods (Boardman *et al.* 2006).

Considering these elasticities within the LCA model is seen as a way to improve the shortcomings of the 1:1 substitution assumption (Ekvall & Weidema 2004; Earles & Halog 2011). On the other hand, Weidema *et al.* (2009) argue that it is reasonable to use elasticity values of 1, providing that the time horizon of the study is long enough so decision makers are able to adapt to changes in price when making investments.

In this PhD work, elasticity values different than 1 have been considered when modeling indirect land use changes. In other words, it was considered that if e.g. 1 kg of barley (or rather carbohydrates providing feed crop) is displaced from Denmark (changed supply), the demand will not remain constant, acknowledging the price changes taking place as a result of this reduced supply.

3. AGRICULTURAL BIOMASS AVAILABLE IN DENMARK

The Renewable Energy Directive (European Union, 2009) defines biomass as “the biodegradable fraction of products, wastes and residues from biological origin […]”.

In a bioenergy perspective, biomass typically stem from agriculture- or forestry-related activities. Over the last few years, the concept of biomass has grown to include organic wastes, here referred to as biowastes (e.g. food waste from households, businesses or service sector; garden waste from households, sport courts or public spaces; and industrial wastes from fish, fruit, sugar, dairy or oil industries, etc.) as well as marine biomass (algae).

As earlier stated, this PhD work focuses on the use of agricultural biomass only, so the other biomass types have not been tackled within this work (although there is scenarios where biowaste and garden waste are involved in Hamelin *et al.* ***V***).

In a Danish context, agricultural biomass can be subdivided into three main categories: energy crops, harvestable crop residues and animal manure.

3.1 Dedicated energy crops

Dedicated energy crops consist of crops that are specifically cultivated for energy purposes. The ideal energy crop has efficient solar energy conversion resulting in high harvestable yield, needs low agrochemical inputs, has low water requirements as well as low moisture level at harvest (Cherubini *et al.* 2009). The chemical composition of the biomass will also influence its suitability for given conversion routes to energy. For example, biomass with low mineral content is preferred for combustion, while biomass with high lignin content may be less suitable for anaerobic digestion (unless a pre-treatment is performed), given its recalcitrance to biological degradation.

Two types of energy crops may be distinguished: annual crops (e.g. wheat, maize, sugar beet) that are sown and harvested annually and perennial crops (e.g. *Miscanthus*, willow) that do not need to be sown every year (i.e. crops having a life cycle of at least 2 years).

Perennial crops are generally well acknowledged as the most efficient and sustainable feedstock available for bioenergy production in temperate regions (Dauber *et al.* 2010; Bessou *et al.* 2011; Valentine *et al.* 2012). Among others, perennial energy crops generally present a more efficient nutrient use than their annual counterpart, which involves lower requirements for annual inputs of fertilizers, and consequently lower environmental impacts related to fertilization (Hamelin *et al.* ***I***). Moreover, in contrast to annual crops whose cultivation tends to accelerate the depletion of soil organic carbon (SOC), perennial energy crops allow for an accumulation of SOC (Hamelin *et al.* ***I***). They generally also present higher yields, involve less soil disturbances due to their longer life-cycle duration, and have a better incidence on biodiversity (Dauber *et al.* 2010). For these reasons, perennials energy crops aroused growing interest in renewable energy strategies worldwide (e.g. Heaton *et al.* 2004; Styles & Jones 2008; Drewer *et al.* 2012) as well as in Denmark (e.g. Gylling *et al.* 2012).

Another important distinction between crops relates to the type of photosynthesis they use to fix the C from the atmosphere: C3 or C4 photosynthesis. The physico-chemical processes and reactions underlying these two photosynthesis types are described in Bolton & Hall (1991) as well as in Zhu *et al.* (2008). In a nutshell, the most important difference between these is that C3 plants (i.e. plants performing C3 photosynthesis) assimilate CO_2 first into a 3-carbon compound, while the first product of photosynthesis of C4 plants is a four-carbon organic acid. As a result, C4 plants can fix more C than C3 plants in conditions of high light and high temperature, resulting in an enhanced light conversion efficiency of ca. 30% (Heaton *et al.* 2008). In fact, according to Heaton *et al.* (2008), the maximum efficiencies with which plants convert solar energy into chemical energy stored as biomass are 6% for C4 plants, and 4.6% for C3 plants.

The potential for energy crop cultivation in Denmark is dependent upon the land available for arable cultivation. Of the total 4.31 Mha of land area in Denmark, 2.79 Mha were used as cropland production (65%), 0.58 Mha as forest (13%), 0.45 Mha as settlement areas[22] (10%), 0.16 Mha as grasslands (4%) and 0.33 Mha (8%) as wetlands and other land types (e.g. sand dunes, beaches, etc.) (Nielsen *et al.* 2011)[23]. As policies have been adopted in order to double the forested areas within the next 80-100 years (Nielsen *et al.* 2011), it appears unlikely that the areas used as forest today could be available for energy crops production. Assuming no changes in the land used as settlement areas, this overview highlights that there is only ca. 2.95 Mha available for energy crop cultivation (i.e. actual cropland and grassland areas). Figure 1 presents the current breakdown of the Danish cropland areas, while land use changes are further discussed in section 4. As it can be seen from Figure 1, half of the current Danish land used as cropland area is used (as of 2012) for wheat and barley cultivation.

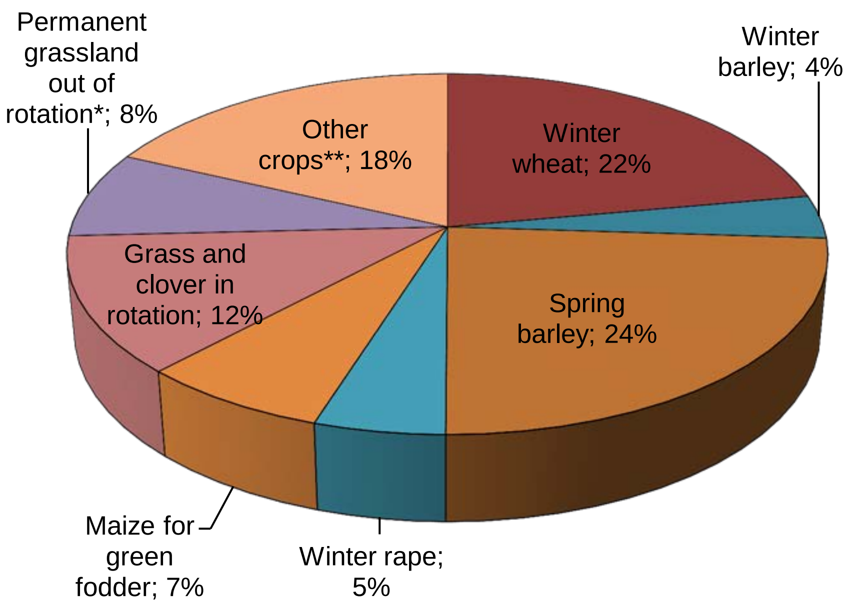

Figure 1. Breakdown of the uses of cropland and grassland areas in Denmark in 2012, based on data from Statistics Denmark (2012c). (*) Portion considered as “cropland”. (**) All crops representing less than 4% were aggregated in this category.

3.2 Harvestable above-ground residues

Crop cultivation generates above- and below-ground biomass, but only the above-ground biomass can be harvested. Above-ground biomass can be divided into primary yield (i.e. the main harvest motivating the cultivation), secondary yield (harvestable residues like straw and tops) and non-harvestable residues (e.g. stubbles, leaves, branches & twigs from woody crops, etc.) (Hamelin *et al.* ***I***). Technically, up to ca. 80% of the secondary yield can be harvested, based on the technologies available today (Birkmose *et al.* 2013)[24].

Although there is growing interest for energy crops species allowing greater secondary yields (e.g. Gylling *et al.* 2012), it must be remembered that the reason why crop cultivation occur at the first place is the demand for the crop itself (primary yield). As such, an increased demand for e.g. straw-based bioenergy will not

[22] Areas with infrastructures, roads, graveyards, sport facilities, etc.
[23] These figures apply for 2009.
[24] According to Gylling *et al.* (2012), there is a potential to increase this further through improving the design of harvesting technologies accordingly.

necessarily mean that more straw-producing crops will be cultivated, as these are primarily produced for something else than straw. Further, when straw-based bioenergy is considered in LCA modeling, the system should be expanded in order to reflect the fate straw would have had otherwise (lost alternative).

From 2006 to 2011, between 4.4 and 5.3 M tonnes of straw DM[25] have been produced yearly in Denmark (Statistics Denmark, 2012d). This straw has been whether left on the field, used for energy, for fodder or for bedding (Figure 2). As it can be seen on Figure 2, the amount of straw used for bedding (11-14% of the straw generated) and fodder (19-23% of the straw generated) appears relatively constant, whereas the amounts left on the field and used for energy are more fluctuant. Figure 2 further shows that most of the straw ends up to be left on the field, where it can be incorporated in the soil. In the light of this, it appears reasonable to consider that if additional straw is used for energy, it is taken away from straw that would have otherwise be plowed down in the soil. This is the lost alternative that was assumed in the case study involving harvesting of straw for energy (Hamelin *et al.* ***V***).

Conservatively considering that only 50%[26] of the straw generated is available for energy production, and considering a biomass LHV average of 18.5 MJ kg^{-1} DM (Haberl *et al.* 2010; Erb *et al.* 2012), the energy potential from straw in Denmark can be roughly estimated to 40-49 PJ y^{-1}. To put this into perspective, straw provided 18.5 to 23.6 PJ y^{-1} of renewable energy in Denmark between 2005 and 2011 (DEA 2012), so there is a potential to approximately double what is already produced today.

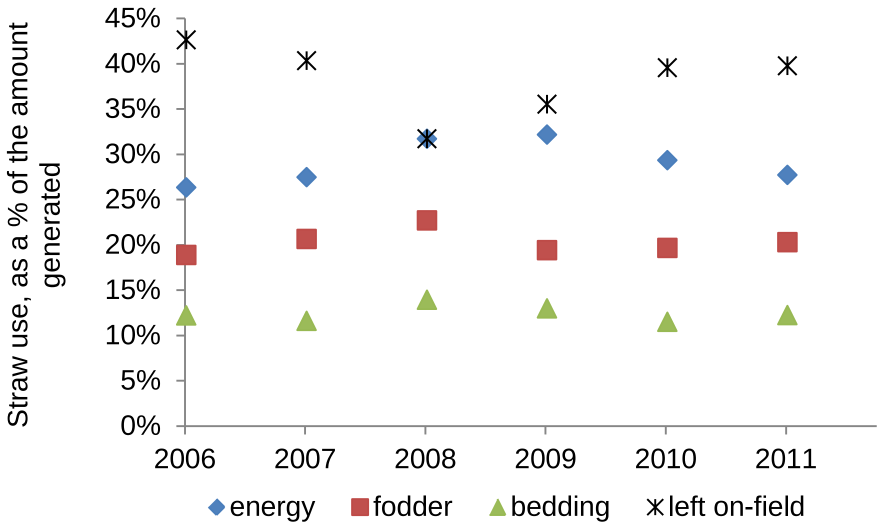

Figure 2. Use of the straw produced in Denmark between 2006 and 2011. Data from Statistics Denmark (2012d)

Harvesting the residues portion that is left on the field is however not as straight-forward as it may seem. There are in fact reasons why these are not harvested, one important reason being that a crop residue cover brings considerable benefits to the soil (e.g. Wilhelm *et al.* 2004; Lal 2005; Johnston *et al.* 2009; Schjønning *et al.* 2009)[27]. In a nutshell, these provide an organic matter input to the soil, which enhance the soil structure through soil aggregation and aggregates stability. Stable aggregates will in turn influence infiltration

[25] Considering 0.85 kg DM per kg fresh matter, based on Møller *et al.* (2000)

[26] This represents the amount already harvested for energy, plus the amount nowadays left on field (the amounts nowadays used for fodder and bedding are not considered as potential). However, it was considered that only 55% of the amount left on-field represents an eventual potential, which is well below the technical harvestable maximal limit of 80% mentioned in Birkmose *et al.* (2013), for example.

[27] It is of course acknowledged that the primary criteria for farmers to harvest the residues or not is market-driven, i.e. if prices of straw rise considerably, it is likely that more farmers will harvest it, even in spite of the lost soil benefits.

of water, soil-water holding capacity, penetration resistance, aeration and bulk density, among others (Wilhelm *et al.* 2004). It is therefore well established that many of the characteristics of highly productive soils relate to their organic matter content, i.e. the soil organic matter[28] (SOM) (Wilhelm *et al.* 2004; Lal 2005). Moreover, a crop residue cover acts as a protection against wind and water (i.e. raindrop) erosion, these decreasing productivity by removing the organic matter-rich topsoil. Declining plant productivity will, in turn, further reduce soil C. In Denmark, however, water erosion risk is a rather minor threat to the agronomic productivity of soils, and although wind erosion has been a significant problem in the past, it is now a minor issue due to the extensive use of hedges as well as winter crops in the regions with sandy soils more prone to such erosion (Schjønning *et al.* 2009).

There is wide evidence that straw removal does decrease SOM and SOC levels (e.g. Wilhelm *et al.* 2004; Thomsen & Christensen 2004; Schjønning *et al.* 2009), but the relation between harvestable residues removal and SOM (or SOC) losses varies strongly from one experiment site to another due to the strong influence of the local conditions on SOM decay (e.g. average temperature, moisture, etc.) as well as local management practices (e.g. tillage). Similarly, not all the C incorporated with the straw or other organic input will result in SOC. Schjønning *et al.* (2009) in fact report that a fixed proportion of the added C is retained in soils, which they quantify, for the Danish soils, as an average of 15% for plant biomass, and 30-40% for animal manure. According to Schjønning *et al.* (2009), the attainable OM storage of a given crop system is essentially defined by the type of crop cultivated (which defines the input of harvestable and not harvestable residues), and by the management practices adopted (e.g. tillage practices, application of manure or mineral fertilizers, residues management, etc.).

Similarly, a variety of responses (i.e. positive and negative) as regards to the impact of residues management on crop yield has been reported across the world. Gabrielle & Gagnaire (2008) report slight decreases (0.5-3%) in wheat primary yield due to straw removal (0.05 to 0.15 t DM ha^{-1} for each tonne of straw removed), which they explain as the lower net mineralization of N in soils resulting from the decrase of SOM incorporation. Similarly, Wilhelm *et al.* (2004) report a loss of 0.13 t DM ha^{-1} (for corn grown in the US) per tonne of corn's crop residues removed. Wilhelm *et al.* (2004) however also report other US field experiments where no differences were observed, and they justify these contradictions in yield response as a result of the contrasting tillage practices employed among experiments. In Denmark, the available experimental evidences cannot confirm the overall conclusion of a yield loss. In fact, the 12-years experiments carried out by Thomsen & Sorensen (2006) showed no significant differences in the yield for spring barley with and without straw incorporation. Similarly, the long-term (18 to 36 years, depending on the soil type) field experiments carried out by Schjønning (2004) showed that while straw incorporation did allow a certain yield increase on sandy soils (ca. 1% per year, observed after 21 years of incorporation only), no effects were observed on clay[29] soils, while it led to a decrease on mixed (or medium) soils[30].

There is therefore an on-going debate about the extent to which harvestable residues can be removed from the fields while maintaining adequate soil quality and productivity. In order to address this issue within this PhD work, soil C changes resulting from different crop management systems (i.e. with and without harvest of the residues) were quantified (Hamelin *et al.* ***I***). This is further described in section 6. In the light of the above-mentioned mitigated evidences on the influence residues management has on crop yield, no attempt were made to establish a relation between straw removal and yield losses in the case studies performed within this PhD work.

[28] Soil organic matter represents the organic constituents in the soil. Although soil organic carbon (SOC) is the major component of soil organic matter (SOM), these two terms should not be seen as synonyms.
[29] To be seen in the light of the Danish context; in Denmark, soils with >10% clay are labeled as clay soils.
[30] On the first location: -0.32% per year, observed after 12 years only; and for the second location, -0.18% per year, observed for the first 13 years, but not changes were observed after that.

3.3 Manure

Denmark is a country with a relatively high agricultural density, both in terms of the livestock and crops produced. As a result, there is, besides the crop residues, a considerable amount of manure available for energy production.

Table 10 presents an estimation of the total amount of manure available in Denmark, per animal and manure types. This theoretical estimation was made based on:

- the livestock population (essentially retrieved from Statistics Denmark, as detailed in Table 10);
- the distribution between the type of manure produced for each animal types (e.g. slurry, deep-litter, etc.)
- the number of days spent in-house (i.e. non-grazing days) (essentially retrieved from the latest national greenhouse gases inventory) and;
- the quantities of manure produced per animal, as presented in the national manure standard 2011 (Poulsen, 2011). These quantities correspond to the amount of manure ex-storage, that is, as manure leaves the long-term storage facilities (section 7).

Based on the data from Table 10, Table 11 presents an estimation of the available energy potential from manure in Denmark (in PJ). These were calculated on the basis of the methane yields of the different manures, and their amount of volatile solids. As shown in Table 11, the total potential would lie between 21 and 30 PJ. This is in line with other national estimates. The Danish Energy Agency, for example, estimates a potential of 26 PJ (DEA, 2010). On the basis of the data from Table 10, Figure 3 further highlights that pig and dairy cattle slurry are the two most important sources of available manure in Denmark (per mass), representing ca. 43% and 35% of the total, respectively[31].

Although manure energy may be recovered through thermal processes, the main (and most obvious) energy conversion pathway used to do so is anaerobic digestion (fermentation process), which is further detailed in section 8. Based on Table 10, it can be seen that the total available manure for energy in Denmark is, on a (wet) mass basis, 34 Mtonne y^{-1} (excluding urine and the amount excreted outdoor). Yet, the amount of manure being digested nowadays in Denmark corresponds to only ca. 5-7% of the manure produced (Birkmose *et al.* 2013). In Denmark, manure-biogas thus represents an underexploited energy potential. However, the amount of renewable energy produced from manure-biogas in Denmark is called to increase significantly, due to the recently launched target to achieve 50% use of manure for biogas by 2020 (Danish Ministry of Food, Agriculture and Fisheries, 2009).

The environmental impacts/benefits related to the use of manure for biogas are discussed in sections 8 and 10.

[31] These percentages are of course different if the proportions are established on the basis of the dry matter content, rather than the mass. Such figures are available in the recent study made by Birkmose *et al.* (2013).

Table 10. Amount of manure available in Denmark. Data for 2010, unless otherwise specified. Amount excreted outdoor not included.

Animal categories	Nb. Animal (A) (head y^{-1})	Number of days in-house[h] (B) (d y^{-1})	Manure type[i] (%) (distribution per animal categories) (C)				Amount ex-storage per animal[j] (t $head^{-1}$ y^{-1}) (D)					Total available manure, estimated (Mtonne y^{-1}) (E) = (A×B×CxD) / (365x10^6)				
			slurry	deep-litter	solid	urine +dong	slurry	deep-litter	solid	urine	dong	slurry	deep-litter	solid	urine[k]	dong[k]
Dairy cattle	**568,202**[a]	347	89%	6%		5%	24.57	15.58		12.93	11.1	11.81	0.50	0.00	0.17	0.15
Non-Dairy Cattle	**1,583,435**															
Bulls & steers, <6 m.	264,036[b]	365		100%				0.96				0	0.25	0	0	0
Bulls & steers, 6 m.-1y	213,458[b]	365	28%	69%		3%	2.94	2.55		1.43	1.97	0.176	0.376	0	0.005	0.006
Bulls & bullocks 1-2 y	38,302[a]	365	28%	69%		3%	2.94	2.55		1.43	1.97	0.032	0.067	0	0.001	0.001
Bulls & bullocks > 2 y	11,629[a]	365	28%	69%		3%	2.94	2.55		1.43	1.97	0.010	0.020	0	0.0002	0.0003
Heifers,<6 m.	314,142[b]	365		100%				1.89				0	0.59	0	0	0
Heifers,6 m.-1 y	312,023[b]	233	71%	29%			6.44	5.52				0.91	0.32	0	0	0
Heifers, 1-2 y	257,048[a]	233	71%	29%			6.44	5.52				0.75	0.26	0	0	0
Heifers, > 2 y	71,710[a]	233	71%	29%			6.44	5.52				0.21	0.07	0	0	0
Suckling cattle	101,087[a]	141	9%	76%		15%	5.5	6.99		2.86	2.61	0.02	0.21	0	0.01	0.01
Swine	**30,045,892**															
Sows	1,116,756[a]	365	90%	7%		1%	4.02	1.79		1.75	0.52	4.04	0.14	0	0.01	0.00
Weaners + piglets	8,254,136[c]	365	98%	2%			0.137	0.027				1.11	0.00	0	0	0
Fattening pigs	20,675,000[d]	365	96%	2%		2%	0.48	0.17		0.33	0.1	9.53	0.07	0	0.07	0.02
Poultry	**112,103,700**															
Broilers	108,204,000[e]	365		100%				0.0016				0	0.17	0	0	0
Laying hens	3,899,700[a]	365	6%	8%	81%		0.0992	0.01105	0.0282			0.02	0.00	0.09	0	0
Turkeys	**200,682**[a]	365		100%				0.0168				0	0.00	0	0	0
Sheep	**159,626**[a]	100		100%				1.16				0	0.05	0	0	0
Horses	**177,500**[f]	182		100%				5.13				0	0.45	0	0	0
Mink	**2,719,600**[g]	365	100%				0.62					1.69	0	0	0	0
TOTAL												**30.30**	**3.58**	**0.09**	**0.27**	**0.19**

[a] Statistics Denmark (2012e)

[b] Statistics Denmark (2012f), quarter 3 + quarter 4
[c] Equal to weaners + piglets – piglets exported. The 2 first data are taken from Statistics Denmark (2012f), and the last data from Landbrug Fødevar (2011).
[d] Equal to slaughtering (total) + export (slaughter pigs + sows) – sows slaughtered and exported. The 2 first data are from Landbrug Fødevar (2011), and the last data is from Statistics Denmark (2012h).
[e] From Statistics Denmark (2012i).
[f] Data from Nielsen *et al.(* 2011), (p. 354), data for 2009. This data is not taken from Statistics Denmark, which does not account for horses from riding schools, or for horses in farms with less than 5 ha.
[g] Statistics Denmark (2012j). Data for 2009.
[h] Nielsen *et al.(* 2011), data for 2009.
[I] Nielsen *et al.(* 2011), data for 2009. When total is not 100%, the difference is the amount excreted outdoor (organic systems).
[j] Poulsen (2011). (Danish manure standards)
[k] Urine and dong roughly distributed as 50% urine, 50% dong.

The breakdown of the available manure for energy per manure types is presented in Figure 3.

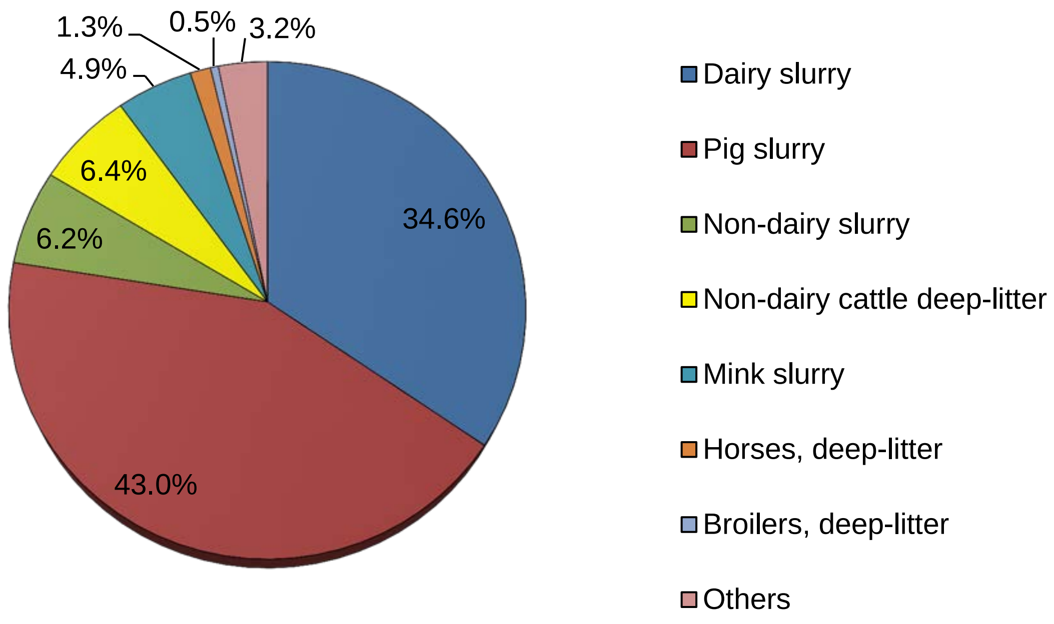

Figure 3. Breakdown of the total available manure in Denmark, per manure type (wet weight).

Table 11. Overview of available energy potential from manure in Denmark (before energy conversion losses)[a]

	1)	2)			3)	4)	5)		6)		7)	
							5) = 2)×3)×4)		6) = 1) × 5)		7) = 6) × 35.2/1000[r]	
Manure type	Amount ex-storage[b]	Methane potential data[c]			VS[j]	TS[n]	Methane potential		Available potential Denmark		Available potential Denmark (in PJ)	
		min	max	unique value			min	max	min	max	min	max
	Mtonne y^{-1}	Nm^3 CH_4 t^{-1} VS			% of TS	t TS t^{-1} manure	Nm^3 CH_4 t^{-1} manure		Nm^3 CH_4 y^{-1} x 10^6		PJ y^{-1}	
Cattle slurry	13.92	150[d]	223.6[e]		80%	0.093[o]	11.2	16.6	155	232	5.5	8.2
Cattle deep litter	2.68	198.7[e]	237[e]		92%[k]	0.3	54.7	65.2	146	175	5.2	6.1
Cattle dung	0.17	100[d]	161[d]		90%[l]	0.228[p]	20.5	33.0	3	5	0.1	0.2
Pig slurry	14.68	232[e]	417[e]		80%	0.066[q]	12.2	22.0	180	323	6.3	11.4
Pig deep litter	0.21			136[f]	80%[m]	0.33[q]	36.0	36.0	8	8	0.3	0.3
Pig dung	0.02	250[d]	403[d]		83%[l]	0.23[q]	47.7	76.9	1	2	0.0	0.1
Poultry slurry	0.02			390[g]	80%	0.12	37.4	37.4	1	1	0.03	0.03
Poultry deep litter (including turkey)	0.18	292[f]	360[g]		80%	0.48	112.1	138.2	20	25	0.7	0.9
Poultry solid	0.09			390[g]	80%	0.4	124.8	124.8	11	11	0.4	0.4
Sheep deep litter	0.05	190[g]	265[h]		80%	0.346	52.6	73.4	3	4	0.1	0.1
Horse deep litter	0.45	265[h]	300[g]		84%	0.26	57.9	65.5	26	30	0.9	1.0
Mink slurry	1.69	350[h]	453[i]		80%	0.065	18.2	23.5	31	40	1.1	1.4
TOTAL											**21**	**30**

[a] Urine not included, nor the amount of manure excreted outdoors.

1)
[b] From Table 10, column E. For "cattle", this is a sum for dairy and all non-dairy cattle species. For "pig", this is a sum of sows, weaners&piglets and fattening pigs. For "poultry", this is a sum for broilers and laying hens.

2)
[c] Values are the biochemical methane potentials (BMP), and not the theoretical biochemical methane potentials (TBMP) (maximum methane producing capacity), unless otherwise indicated. "Min" and "max" indicate the minimum and maximum BMP values found in the literature for each relevant category.
[d] Møller *et al.* (2004). In the pig dung case, the minimum value is for sows, and the maximum for fattening pigs.
[e] Triolo *et al.* (2011). In the pig slurry case, the minimum value is for sow manure, and the maximum value for piglets manure.
[f] Based on an average from AERBIOM (2009) and data from the Baltic Manure project (see note j: the data used are those of Germany, Sweden and Estonia).
[g] IPCC (2006a). The values from this reference are reported as TBMP.
[h] Jørgensen (2009).
[i] Triolo *et al. (in press)*

3-4)
[j] Values from Baltic manure project (www.balticmanure.eu), unless otherwise specified.
[k] Triolo *et al.* (2011)
[l] Møller *et al.* (2004)

[m] Assumption
[n] Poulsen (2011). (Danish manure standards)
[o] Value for dairy
[p] Value for suckling cows
[q] Value for fattening pigs

7)
[r] The LHV of methane, at normal conditions (0°C, 100 kPa), is taken as 35.2 MJ/Nm^3.

4. DIRECT AND INDIRECT LAND USE CHANGES

4.1 Land is limited

Although agricultural biomass is a renewable energy resource, it is not unlimited in supply. In the case of straw and manure, the amounts produced are limited by other production activities, namely crop and animal production, as earlier discussed (section 2.2). The production of energy crops, on the other hand, is dependent upon the land available to grow it, and thus limited by the amount of land available for cultivation (among others limitations).

Land is in fact a finite resource. According to FAOSTAT (2012c), there is 13 Gha of land area on Earth (this does not include the area under inland water bodies). Of this 13 Gha, there is (for year 2009, the latest year for which, at the time of writing, data are available in FAOSTAT):

- 4.89 Gha of agricultural land
  - 1.53 Gha arable land;
  - 3.36 Gha permanent meadows and pastures.
- 4.04 Gha of forest[32]
  - 3.76 Gha natural forest;
  - 0.28 Gha plantations.
- 4.09 Gha of other land[33]
  - 2.50 Gha of uncultivable land such as tundra, ice and desert;
  - 1.59 Gha built-up land, savannahs and alike areas, and any other land not included in the above.

From this overview, it can be seen that there is a maximum of approximately 3.76 Gha of land not already used by humans that could potentially be cultivated for growing energy crops, together with part of the 3.64 Gha currently used for pasture/meadows and plantations.

Yet, only part of this land is suitable for agricultural cultivation in practice. In order to establish a quantitative estimate of the maximal available land that could be suitable for new cultivation (maximal biophysical potential), Ramankutty *et al.* (2002) mapped the world as 0.5-degree resolution grid cells (latitude × longitude) and developed an index of land suitability representing the probability that each particular grid cell is cultivated. To determine if the characteristics of a particular grid cell allowed for cultivation, Ramankutty *et al.* (2002) considered two main parameters, namely climate and soil properties, which they argue to represent the major constraints for cultivation, on a global scale. The main conclusion from this study is that the total extent of suitable land for cropland cultivation is 4.10 Gha[34] (this includes the portion already under cultivation), most of this potential being located in tropical Africa and northern South America.

This indicates that the current cropland area can at maximum be doubled. Yet, this is to be seen as a maximal theoretical figure, as the 4.10 Gha figure of Ramankutty *et al.* (2002) was derived without considering the influence of topography, nor considering the zones that are today protected (conservation zones). Further, socio-economic aspects are not considered either in this estimate; the suitable land located in conflict or unstable areas may in practice find few investors/farmers willing to implement a plantation on

[32] According to FAO (2010), this consists of 36% primary forest (with native species), 57% regenerated forest and 7% plantations. According to Kampman *et al.* (2008), about 78% of these plantations are "productive" plantation (i.e. established for wood and fibre production), while 22% are under protection (conservation areas).
[33] According to FAOSTAT (2012d),"other land" includes the land that is not under water bodies and that is not classified as agricultural land or forest area. It includes, among other, barren land and built-up areas. The breakdown provided here is based on the data available in Kok *et al.* (2008).
[34] Or 4.76 Gha under a climate change scenario for 2070-99

these. Moreover, RFA (2008) reports other studies where lower estimates have been derived, with respect to the suitable additional cultivable land available: one by IIASA[35] with a result of 0.7 to 1.2 additional Gha, and one by EEA[36] with an additional 0.05 to 0.4 Gha only, the lower estimate considering zero conversion of grassland to cropland.

The available additional cultivable land is, thus, limited. Yet, increased bioenergy is only one out of several other competing demands for this new cultivable land potential. Foley *et al.* (2011) in fact report that besides bioenergy, crop production needs to roughly double just to keep pace with projected demands from population growth and dietary changes towards more meat consumption in the so-called transition countries. For example, recent studies (e.g. FAO 2008; Nonhebel 2012) forecast that the demand of cereals, only for food and feed needs (i.e. excluding bioenergy), will increase by ca. 50% within the next 20 years.

In a LCA perspective, acknowledging that the extent of available cultivable land is limited implies that any process requiring the use of additional cultivable land has a “lost alternative” cost related to it, which must be modeled in the LCA. This lost alternative may be, among other possibilities, the displacement of another crop cultivation system, the displacement of grazing land or the displacement of natural ecosystems, as further discussed in section 4.3. Such displacements, or changes in the use of land, are termed as “land use changes”, and are often distinguished as “direct” and “indirect” land use changes, as further described below.

These land use changes are a concern because of the considerable environmental cost they could involve, particularly if they lead to the expansion of crop production in carbon-rich ecosystems, causing the release of carbon that was stored in these ecosystems over long periods of time (e.g. Gibbs *et al.* 2008). In fact, as highlighted in recent studies (e.g. Ramankutty *et al.* 2002; Morton *et al.* 2006; Kløverpris 2008; Gibbs *et al.* 2008), biomes like tropical or temperate forests are those that are likely to be the first converted following an increase demand for crops, these being the biomes where the frontier between agriculture and nature is already moving. This consequence is often termed “C debt”, reflecting the number of years of bioenergy offsetting fossil fuels that are needed before to balance the amount of C released from the conversion of natural ecosystems to agriculture.

4.2 Direct land use change (DLUC)

As explained in section 2.6, spring barley (Tonini *et al.* ***II***) and maize (Hamelin *et al.* ***V***) were the short- and long-term marginal crops considered for Denmark in this study, i.e. the crops that are displaced by more land being used to grow energy crops in Denmark. Cultivating a given energy crop instead of spring barley or maize thus represents the direct land use change modeled in this PhD work, i.e. the “immediate” change in the land use allocation occurring as a result of increased bioenergy in Denmark. For the LCA model, the interest lies in the environmental impacts deriving from using the land to cultivate a given energy crop instead of the displaced crop.

In the case studies performed in this PhD work (e.g. Tonini *et al.* ***II***; Hamelin *et al.* ***V***), the environmental consequences related to DLUC were modeled based on the inventory data provided in Hamelin *et al.* (***I***).

4.3 Indirect land use change (ILUC)

As more land is used for bioenergy in Denmark, the resulting drop in supply of marginal food/feed crop from Denmark (here spring barley or maize) will, based on the consequential LCA logic as well as on recent studies (e.g. Kløverpris 2008; Searchinger *et al.* 2008), cause a relative increase in agricultural prices, which

[35] International Institute for Applied System Analysis
[36] European Environment Agency

then provides incentives to increase the production elsewhere[37]. Such responding increase may be achieved through three main mechanisms:

(i) Transformation of non-cultivated area (nature) to cropland, also referred to as land expansion (or new land cultivation).
(ii) Increased yield per land area, also referred to as intensification
(iii) Displacement(s) of other crops and livestock

While (i) and (ii) represents ultimate responses, (iii) is to be seen as an intermediate response leading, through cascading effects, to final intensification and/or land expansion somewhere on the planet. In fact, once food/feed is displaced in one location, it may be replaced by a reacting feedstock in another location, which itself may displace another feedstock, and so forth until replacement is only achieved by intensification and land expansion. This chain of events leading to final net intensification and/or expansion will be determined by crop substitutability and responses to changing prices, among others.

When "virgin" land is converted to agriculture, not only is the C from this land's vegetation and soil released, but this vegetation would, in many cases, have continued to absorb C[38], which would reduce the net CO_2 in the atmospheric pool. The difference between the amount of C that would have been absorbed by this vegetation and the amount of C that is absorbed by the new agricultural production is referred to as the foregone sequestration capacity (e.g. Searchinger *et al.* 2008).

In brief, the overall ILUC response to increased bioenergy in Denmark consists of a series of 5 main impacts, all of which carries an environmental cost:

(i) Arable land expansion (conversion of nature to agriculture) (ALE)
(ii) Intensification (I)
(iii) Displacement of crops and livestock (DI)
(iv) Cultivation of the reacting crops on the land converted to agriculture (RC)
(v) Foregone sequestration capacity (FS)

In other words, the overall environmental impacts from ILUC, for a given impact category (ic), is the sum of the impacts from all above terms:

$$\textit{Environmental impacts } ILUC_{ic} = ALE_{ic} + I_{ic} + DI_{ic} + RC_{ic} + FS_{ic} \qquad \text{(Equation 1)}$$

All these points are addressed in the sections below. It could be argued that RC and FS could be merged and viewed as a form of direct land use changes at the location where conversion occurs, i.e. reflecting the consequences of cultivating arable crops instead of having the natural vegetation to grow on the land. Here, these are however addressed as two separate points.

In most ILUC studies carried out so far, only ALE is quantified, in terms of environmental impacts. Likewise, this PhD work only considered this impact when quantifying the global warming potential (GWP) of ILUC (Tonini *et al.* ***II***). In Hamelin *et al.* (***V***), the impact of RC was also considered.

In this PhD work, the point of departure for ILUC to occur is the increased cultivation of bioenergy plantations in Denmark. These plantations, typically, would occur over several years, e.g. between 10 to 20 years. When performing LCA, it is often needed to express the emissions occurring over a certain period of time on an

[37] In the light of section 2.7, it must be emphasized that not necessarily all barley/maize displaced from Denmark will be replaced. In fact, as displacing these crops will lead to higher prices for these commodities, it is often forecasted that the demand for these will simply decrease, so part of the displaced feedstock is never replaced (e.g. Hertel *et al.* 2010; Laborde 2011).

[38] Although this is particularly true for re-growing forests, it may not necessarily be the case for "mature" forests, where little C is sequestered in the soil and vegetation.

annual basis, in order to aggregate these emissions with the other (annual) flows involved in the system. In the case of ILUC, it is common practice to distribute the emissions over 20 years (European studies[39]) or 30 years (US studies[40]). This practice, where ILUC emissions are allocated to a certain volume of bioenergy, is generally referred to as “annualization”, or “amortization” of the ILUC emissions. Although there are some debates over the relevance of this practice (e.g. Schmidt *et al.* 2012; Kløverpris & Mueller 2013), it has been, within this PhD work, simply accepted as a common standard to deal with ILUC emissions. ILUC emissions, as well as other emissions occurring over a certain time frame (e.g. soil C changes) have thus been annualized over a period of 20 years, unless otherwise stated.

4.3.1 Land expansion

The land expansion impact corresponds to the environmental consequences of converting land nowadays not used for crop cultivation to cropland, as a result of the induced demand for the displaced food/feed crops (here barley or maize). To quantify this impact, it is necessary to:

i. Identify how much land is converted, where it is converted and which types of land are converted (biome types);
ii. Estimate, for all converted biomes, the releases of C from the vegetation and soil to the atmosphere.

So far, most studies attempting to quantify point (i) above used econometric models to this end (e.g. Kløverpris 2008; Searchinger *et al.* 2008; Edwards *et al.* 2010; Tyner *et al.* 2010; Laborde 2011). The reason for this is due to the very nature of the ILUC process: changes in land use result in changes in crop supply that are transmitted across global markets linked by commodity substitutability and competition for land through numerous interactions. To cope with these, sophisticated models allowing to represent the world crop markets become essential.

Econometric models may be based on partial or general equilibrium. Partial equilibrium models consist of a very detailed representation of a single sector of the economy (e.g. agriculture, energy). In contrast, general equilibrium models aim at modeling the whole world economy, but fewer details are then considered. A comprehensive overview of partial and general equilibrium models that can be used to estimate ILUC is given in Witzke *et al.* (2008), while a summarized overview is provided in Prins *et al.* (2010). The strength of these models is that they combine into one single modeling framework both economic and biophysical/agricultural considerations (e.g. crop yields, land suitability for given crop cultivation, etc.).

In this study, the approach adopted was to use the output of a study where an econometric model was applied, and to link it to one ha of marginal crop displaced in Denmark. To this end, the study performed by Kløverpris (2008) was used, where the ILUC consequences in terms of points (i) above are identified, on the basis of a modified version of the general equilibrium GTAP model (Hertel 1999), for a marginal increase in wheat consumption in 4 different countries, including Denmark.

In Tonini *et al.* (***II***) as well as in Hamelin *et al.* (***V***), the output results of Kløverpris (2008) for Denmark have thus been used as a proxy to estimate how much land is converted (due to the increased spring barley/maize demand generated by the displaced spring barley/maize from Denmark), where in the world and from which biome (point i above). More details explaining how this procedure was performed are available in the SI of Tonini *et al.* (***II***) (Appendix B).

In order to quantify the releases of C due to land conversion (point ii above), the soil and vegetation carbon data from the Woods Hole Research Centre, as published in the “supporting online material” of Searchinger *et al.* 2008 have been used. From this database, the amount of C in the soil and vegetation of all affected biomes (point i) were extracted. This allowed to calculate the CO_2 emitted during land conversion. This

[39] Based on the IPCC (2006b) guidelines as well as on the Renewable Energy Directive, itself based on the IPCC guidelines.
[40] Based on the publication from Searchinger *et al.* (2008).

calculation was performed based on the methodology reported in Müller-Wenk & Brandão (2010). Based on this methodology, it was considered that:

- 25% of the C in the soil was released as CO_2 for all types of land use conversion, except when forests were converted to grassland, where 0% was released;

- 100% of the C in vegetation was released as CO_2 for all forest types as well as for tropical grassland conversions[41], while 0% was released for the remaining biome types (e.g. shrub land, non-tropical grassland, chaparral).

An example of the application of this approach is presented in Table 12. It lead to a land expansion CO_2 figure of 310±170 t CO_2 ha^{-1} (Tonini *et al.* ***II***) or 357±195 t CO_2 ha^{-1} (Hamelin *et al.* ***V***), depending on the yield considered for the crop displaced[42].

[41] This is to be seen as a simplifying assumption (personal communication with Miguel Brandão, ILCA, January 2013, and with David Laborde, IFPRI, February 2013). However, from the data of Earles *et al.* (2012), whom detailed, for 169 countries, the fate of the above-ground residues when forest are cleared, it can be seen that even after 100 years, it is not exactly 100% of the C that is returned to the atmosphere, although the gap is negligible in most cases.

[42] In fact, the greater the yield of the crop displaced, the greater the ILUC will be, as more biomass is then displaced (and thus have to be replaced).

Table 12. Estimation of the ILUC CO_2 impact[a] (adapted from Tonini *et al.* ***II***)

Results from Kløverpris (2008)				Woods Hole database (Searchinger *et al.* 2008)		Müller-Wenk & Brandão (2010)	Own calculations	
Biomes converted[b]	Conversion to[c]	Region[d]	m^2 t^{-1} wheat[e] (A)	C in vegetation (t ha^{-1}) (B)	C in soil (t ha^{-1}) (C)	CO_2-C lost (t C t^{-1} wheat)[f] (D)	CO_2 lost (t CO_2 t^{-1} wheat) (E)	CO_2 lost per ha displaced (t CO_2 ha^{-1})[g] (F)
Savannah (taken as shrub land)	C	xss	140 ± 86	4.6	30	0.11 ± 0.06	0.39 ± 0.24	2.2 ± 1.3
African tropical evergreen forest (taken as tropical rain forest)	C	xss	140 ± 86	130	190	2.5 ± 1.5	9.1 ± 5.5	52 ± 31
Open shrubland (taken as shrub land)	G	xss	81 ± 49	4.6	30	0.06 ± 0.04	0.22 ± 0.13	1.3 ± 0.8
Temperate evergreen forest	C	xeu15	57 ± 34	160	130	1.1 ± 0.7	4.0 ± 2.4	23 ± 14
Temperate deciduous forest	C	xeu15	57 ± 34	120	130	0.87 ± 0.52	3.2 ± 1.9	18± 11
Dense shrub land (taken as temperate grassland)	C-G	xeu15	250 ± 150	7.0	190	1.2 ± 0.7	4.3 ± 2.6	24 ± 15
Tropical evergreen forest	C	bra	180 ± 70	200	98	4.0 ± 1.6	15 ± 6	83 ± 33
Savannah (taken as grassland)	G	bra	41 ± 16	10	42	0.04 ± 0.02	0.16 ± 0.06	0.91 ± 0.36
Grassland/steppe (taken as temperate grassland)	C	xsu	91 ± 55	10	190	0.43 ± 0.26	1.6 ± 0.9	9.0 ± 5.4
Temperate evergreen forest	G	xsu	45 ± 27	160	130	0.88 ± 0.43	3.2 ± 1.6	18.3 ± 9.1
Temperate deciduous forest	G	xsu	45 ± 27	140	130	0.76 ± 0.37	2.8 ± 1.3	16 ± 8
Savannah (taken as tropical grassland)	C	aus	110 ± 64	18	42	0.31 ± 0.18	1.1 ± 0.7	6.4 ± 3.8
Open shrubland + grassland/steppe (taken as tropical grassland)	G	aus	37 ± 22	18	42	0.11 ± 0.06	0.39 ± 0.23	2.2 ± 1.3
Boreal deciduous forest (taken as temperate deciduous forest)	C	can	97 ± 58	140	130	1.6 ± 1.0	6.0 ± 3.6	34 ± 20
Boreal evergreen forest (taken as temperate evergreen forest)	G	can	10 ± 6	160	130	0.16 ± 0.10	0.59 ± 0.35	3.3 ± 2.0
Grassland/steppe (taken as grassland)	C	xla	35 ± 21	10	42	0.04 ± 0.02	0.14 ± 0.08	0.77 ± 0.46
Tropical evergreen forest	C	xla	35 ± 21	200	98	0.79 ± 0.48	2.9 ± 1.7	17 ± 10
Savannah + dense shrub land (taken as grassland)	G	xla	16 ± 10	10	42	0.02 ± 0.01	0.063 ± 0.038	0.36 ± 0.22
Open shrub land (taken as chaparral)	G	usa	68 ± 41	40	80	0.14 ± 0.08	0.50 ± 0.30	2.8 ± 1.7
TOTAL	-	-	1500 ± 880	-	-	15 ± 8	54 ± 30	310 ± 170

[a] Eventual inconsistencies due to rounding (numbers are reported with 2 significant digits).
[b] Indicated biomes are as in Kløverpris (2008). When the biomes mentioned in Kløverpris (2008) did not figure in the biomes from the Woods Hole Research Centre data (Searchinger *et al.* 2008), an equivalent was considered, which is indicated between parentheses, when it applies.
[c] C: cropland; G: Grassland. For dense shrubland, the conversion is 46% to cropland, 54% to grassland.
[d] With xss: Sub-Saharan Africa, excluding Botswana, Lesotho, Namibia, South Africa and Swaziland; xeu15: EU-15, excluding Denmark; bra: Brazil; xsu: Former Soviet Union, excluding the Baltic States; aus: Australia; can: Canada; xla: South America, excluding Brazil and Peru; usa: United States. As indicated in Kløverpris (2008), this aggregation covers 92% of the total net expansion.
[e] The maximal and minimal range are based on the qualitative description of the uncertainty related to the biomes conversion results made by Kløverpris (2008). The levels identified as “very good”, “good” and “moderate” were considered as an uncertainty of ±20%, 40% and 60%, respectively.
[f] Calculated as: (D) = (A) × ha/10000 m^2 × [(% released from soil × (B)) + (% released from vegetation × (C))]. The % of C releases from soil and vegetation are as indicated in the text.
[g] The conversion per ha is made considering that it is 1 ha of spring barley that is initially displaced, with a yield of 4.85 t DM ha^{-1} and a DM content of 85% of the crop fresh matter.

Most studies available to date where ILUC is taken into account aimed to calculate the land expansion impact of large-scale biofuel policies (e.g. Europe, US). Their results are typically reported in terms of CO_2 emitted per unit of primary energy in the fuel (i.e. prior to combustion). In Table 13, an overview of results from these studies is summarized, and compared with the results obtained in the present study, per unit of energy. The same exercise has been performed in Table 14, where the few available results per hectare from other studies are compared with those obtained in this study. This comparison, however, should be interpreted carefully, as the land expansion CO_2 values obtained in biofuel studies aggregate a variety of effects that do not apply here (e.g. when biofuels by-products are used for animal feed, this feed is not produced anymore, which reduces the overall ILUC modeled by these studies). Nevertheless, it can be seen from the overview below that the results obtained in this PhD work are in the range of those derived by previous studies, both per energy intensity (Table 13) and per hectare (Table 14).

Table 13. Overview of land expansion figures reported in recent studies, per unit of energy produced.

Scenario modeled	**ILUC (ha expanded TJ^{-1})**	**ILUC, annualized[a] (g CO_2 eq. MJ^{-1} y^{-1})**	**ILUC, not annualized (g CO_2 eq. MJ^{-1})[f]**	**Ref.[g]**
Biodiesel				
Soy (US demand), FAPRI-CARD model	2.0[d]	30 – 99[b]	600 – 1980	[1]
Soy (EU demand), MIRAGE-BIOF model	4.0[c]	56 - 57	1120 - 1140	[2]
Rapeseed (EU demand), MIRAGE-BIOF model	4.0[c]	55 - 56	1100 - 1120	[2]
Sunflower (EU demand), MIRAGE-BIOF model	5.0[c]	54 - 55	1080 – 1100	[2]
Palm oil (EU demand), GTAP model	1.9	47	940	[3]
Palm oil (EU demand), LEITAP model	10	325[c]	6500	[3]
Mix feedstock (US demand), AGLINK model	5.7	40[c]	800	[3]
Mix feedstock (EU demand), GTAP model	9.1	57	1140	[3]
Mix feedstock (EU demand), AGLINK model	5.5	40[c]	800	[3]
Mix feedstock (EU demand), FAPRI-CARD model	11	100[c]	2000	[3]
Mix feedstock (demand from Germany), LEITAP model	46	350[c]	7000	[3]
Mix for EU consumption (ca. 86% rapeseed, 12% soy, 2% palm oil), based on historical land conversion data	5.7 – 11	30 - 204	600 - 4080	[4]
Average[e] [min-max]	***9 [2-46]***	***109 [30-350]***	***2180 [600-7000]***	
Bioethanol				
Wheat (EU demand), GTAP model	19	155	3100	[3]
Wheat (EU demand), FAPRI-CARD model	9.3	75[c]	1500	[3]
Wheat (EU demand), IMPACT model	5.3	40[c]	800	[3]
Wheat (EU demand), AGLINK model	14	100[c]	2000	[3]
Wheat (demand from France), LEITAP model	17	145[c]	2900	[3]
Wheat (EU demand), MIRAGE-BIOF model	1.4[c]	13 - 14	260 – 280	[2]
Wheat (US demand), IMPACT model	5.3	40[c]	800	[3]
Coarse grains (EU demand), IMPACT	2.9	25[c]	500	[3]
Coarse grains (US demand), GTAP model	4.1	62	1240	[3]
Coarse grains (US demand), AGLINK	12	85[c]	1700	[3]
Corn (US demand), LEITAP model	21	150[c]	3000	[3]
Corn (US demand), IMPACT model	2.6	25[c]	500	[3]
Corn (US demand), FAPRI-CARD model	0.61[d]	33 – 60[b]	660 – 1200	[1]
Corn (US demand), GTAP model	3.5	22 - 135	440 – 2700	[5]
Corn (US demand), GTAP model	1.5[d]	21 – 35[b]	420 – 700	[6]
Corn (US demand), FAPRI-CARD model	9.2[d]	156[b]	3120	[7]
Mix for EU consumption (ca. 55% wheat, 35% sugar beet, 10% sugar cane), based on historical land conversion data	4.3 – 6.9	26 - 154	520 – 3080	[4]
Switchgrass (US demand), FAPRI-CARD model	Not given	167[b]	3340	[7]
Switchgrass (US demand), FAPRI-CARD model	0.20[d]	14 – 31[b]	280 – 620	[1]
Cellulosic biofuel (at global scope) to stabilize CO_2 at 550 ppm, EPPA model	Not given	278 – 285[b]	5560 – 5700	[8]
Sugar beet (EU demand), MIRAGE-BIOF model	0.4[c]	5 - 7	100 – 140	[2]
Sugar cane (EU demand), MIRAGE-BIOF model	1.5[c]	15 - 20	300 – 400	[2]
Sugar cane (EU demand), AGLINK model	3.1	25[c]	500	[3]
Average[e] [min-max]	***7 [0.2-21]***	***80 [5-282]***	***1600 [100-5640]***	
Solid and gaseous biomass for heat and electricity, this study				
Ryegrass (this study, different conversion technologies)	6 – 8[h]	91 – 129[h]	1820 – 2580	[9]
Miscanthus (this study, different conversion technologies)	6 – 8[h]	91 – 129[h]	1820 – 2580	[9]
Willow (this study, different conversion technologies)	5 – 7[h]	70 – 103[h]	1400 – 2060	[9]
Maize silage (this study, anaerobic co-digestion with manure)	14	106	2120	[10]
Average[e] [min-max]	***9 [5-14]***	***103 [70-129]***	***2060 [1400-2580]***	

[a] 20 years annualization, as explained in section 4.3.
[b] The original result was annualized over a time horizon of 30 years; the result presented here is the converted value (for a 20 years annualization period).
[c] Value taken from a graph.
[d] Considering, in the conversion, a lower heating value of 21 MJ per L for ethanol (European Union 2009).
[e] In cases with a range, the median was used to compute the average.
[f] Annualized value times 20 years.

[g] Reference : 1. USEPA (2010); 2. Laborde (2011); 3. Edwards *et al.* (2010); 4. Overmars *et al.* (2011); 5. Hertel (2010); 6. Tyner *et al.* 2010; 7. Searchinger *et al.* 2008; 8. Melillo *et al.* (2009); 9. Tonini *et al.* (***II***); 10. Hamelin *et al.* (***V***).
[h] For the average ILUC, with the range representing the different conversion technologies.

Table 14. Overview of land expansion values reported in recent studies, per ha displaced

Scenario modeled	ILUC (t CO_2 ha^{-1})	Reference
Increased bioenergy in Denmark[a]	310 (140 - 480)	Tonini *et al.* (***II***)
Increased bioenergy in Denmark[a]	357 (162 - 552)	Hamelin *et al.* (***V***)
Increased bioenergy in the EU	200 – 550[b]	Laborde (2011)
Increased bioenergy in the US	351	Searchinger *et al.* (2008)
Increased bioenergy in Germany	400[c]	Sterner & Fritsche (2011)
Average, excluding results from this study[d] [min-max]	***375 [200-550]***	

[a] Average value with range indicated between parenthesis
[b] Values taken from a graph.
[c] This value corresponds to the maximal ILUC, i.e. where the response is met by land expansion only.
[d] In cases with a range, the median was used to compute the average.

4.3.2 Displacement of crops and livestock

Displacement of crops and livestock, is a form of direct land use changes occurring throughout the chain of events leading to intensification and/or expansion. Although it has not been addressed within this PhD work, the principles to be applied are the same as when addressing direct land use changes: i.e. the environmental consequences represent the difference between the new use of land versus the displaced use of land.

While most ILUC studies based on econometric models do quantify the “physical” displacement effect (in terms of hectares of crop displaced, biomes where displacement occurs, crops replacing the crops displaced, yield of the replacing and displaced crops, etc.), none quantified, to author’s knowledge, the environmental impacts related to it. The main challenge for doing so is the availability of quality datasets (particularly soil C changes), such as those presented in Hamelin *et al.* (***I***), for all crop and biome systems where displacement-replacement occurs.

4.3.3 Intensification

Intensification refers to the increase of crop yields as a response to increasing crop market price. Recent studies on biofuels or increased crop consumption involving economical modeling indicated that the share of the intensification response in replacing the displaced biomass is likely to be of at least 15% (Kløverpris 2008; Marelli *et al.* 2011) and may potentially be as high as 70% (Marelli *et al.* 2011).

In the case studies performed within this PhD work, like in most available studies, the contribution of intensification to the total environmental implications of ILUC was not considered. However, the main characteristics of intensification are discussed below. Environmental implications, and potential methodologies to quantify them, are discussed in section 10.4.4.

Intensification may be achieved through three main pathways:

- Input-driven pathway: this refers to any yield increases obtained through changes in farm inputs (e.g. fertilizers, pesticides, irrigation, etc.). The increases in yield obtained this way may however be reversible.
- Innovation-driven pathway: this refers to any yield increases obtained through technological development (e.g. harvesting technologies allowing to recover more biomass, plant breeding, etc.), and is seen as a more permanent effect (Marelli *et al.*, 2011). However, a lag of ca. 20 years is likely before research and development activities actually translate into yield increases (Edwards *et al.*, 2010).
- Multi-cropping pathway: this consists to grow more than one crop on the same hectare of land for a given year, which in some countries allows a harvest all year-round. This currently represents 18%

of the world's cropland, and higher crop prices can be envisioned to increase the profitability of this practice (Marelli *et al.* 2011).

While input-driven and multi-cropping intensification are directly driven by crop prices, innovation-driven intensification is dependent upon public and private research investments, and as such is more difficult to predict (Kløverpris 2008). However, it is well acknowledged as the most sustainable way to increase the yields, especially in developing countries (Marelli *et al.* 2011; Tilman *et al.* 2011)

According to Tilman *et al.* (2002), multi-cropping has the potential to improve pest control and increase nutrient- and water- use efficiency, besides contributing to increase the amount of biomass harvested annually. The extent to which producers will decide to opt for multi-cropping is however governed by the difference between the production costs and the value of the production. This also applies for input-driven intensification, which is further characterized by the so-called "diminishing returns", meaning that for each additional unit of input (e.g. fertilizer) applied, the magnitude of the additional yield becomes lower and lower, until it becomes practically negligible.

4.3.4 Foregone sequestration

Foregone sequestration has not been accounted for in the case studies performed within this PhD (Hamelin *et al.* ***V***; Tonini *et al.* ***II***). Using the same background data as in Table 12 (itself from Tonini *et al.* ***II***), foregone sequestration can be estimated to 21 ± 13 t CO_2 eq. per hectare displaced in Denmark. This is detailed in Table 15. This figure, which represents the amount of C that would have been absorbed by the natural ecosystems if land use changes would not have happened, is not as significant, magnitude-wise, as the releases occurring due to land expansion (310 ± 170 t CO_2 eq. ha^{-1} displaced). One reason for this is that, based on the data from the Woods Hole Research Centre C database, very little C would have been absorbed by the mature forests that are converted (this value is even zero for the tropical evergreen forest in Brazil and Latin America), while no foregone sequestration (at all!) is considered for biomes under tropical savannah and shrub lands. Further, it should be highlighted that only CO_2 absorption from the atmosphere is accounted for. Other C fluxes affected as a result of land conversion, like for example the uptake of atmospheric CH_4 from soils (Robertson *et al.* 2000; Smith *et al.* 2000: section 6.4), are not considered in this estimate.

Table 15. Estimation of the CO_2 impact for 20 years of foregone sequestration, for the case study presented in Tonini *et al.* (***II***)[a]

Results from Kløverpris (2008)				Woods Hole database[f]	Own calculations
Biomes converted[b]	Conversion to[c]	Region[d]	$m^2\ t^{-1}$ wheat[e]	20 years of uptake from natural vegetation (t ha^{-1} of biome converted)	20 years of foregone CO_2 sequestration (t CO_2 ha^{-1} displaced)
Savanna (taken as shrub land)	C	xss	140 ± 86	0	0 ± 0
African tropical evergreen forest (taken as tropical rain forest)	C	xss	140 ± 86	1.8	0.54 ± 0.33
Open shrubland (taken as shrub land)	G	xss	81 ± 49	0	0 ± 0
Temperate evergreen forest	C	xeu15	57 ± 34	42	5.0 ± 3.0
Temperate deciduous forest	C	xeu15	57 ± 34	37	4.4 ± 2.6
Dense shrub land (taken as temperate grassland)	C-G	xeu15	250 ± 150	4.7	2.4 ± 1.4
Tropical evergreen forest	C	bra	180 ± 70	0	0 ± 0
Savanna (taken as grassland)	G	bra	41 ± 16	0	0 ± 0
Grassland/steppe (taken as temperate grassland)	C	xsu	91 ± 55	4.7	0.89 ± 0.53
Temperate evergreen forest	G	xsu	45 ± 27	42	3.9 ± 2.4
Temperate deciduous forest	G	xsu	45 ± 27	37	3.5 ± 2.1
Savanna (taken as tropical grassland)	C	aus	110 ± 64	0	0 ± 0
Open shrubland + grassland/steppe (taken as tropical grassland)	G	aus	37 ± 22	0	0 ± 0
Boreal deciduous forest (taken as temperate deciduous forest)	C	can	97 ± 58	1.3	0.26 ± 0.16
Boreal evergreen forest (taken as temperate evergreen forest)	G	can	10 ± 6	9.9	0.21 ± 0.12
Grassland/steppe (taken as grassland)	C	xla	35 ± 21	0	0 ± 0
Tropical evergreen forest	C	xla	35 ± 21	0	0 ± 0
Savanna + dense shrub land (taken as grassland)	G	xla	16 ± 10	0	0 ± 0
Open shrub land (taken as chaparral)	G	usa	68 ± 41	0	0 ± 0
TOTAL	-	-	1500 ± 880	-	21 ± 13

[a] Eventual inconsistencies due to rounding (numbers are reported with 2 significant digits).
[b] Indicated biomes are as in Kløverpris (2008). When the biomes mentioned in Kløverpris (2008) did not figure in the biomes from the Woods Hole Research Centre data (Searchinger *et al.* 2008), an equivalent was considered, which is indicated between parentheses, when it applies.
[c] C: cropland; G: Grassland. For dense shrubland, the conversion is 46% to cropland, 54% to grassland.
[d] With xss: Sub-Saharan Africa, excluding Botswana, Lesotho, Namibia, South Africa and Swaziland; xeu15: EU-15, excluding Denmark; bra: Brazil; xsu: Former Soviet Union, excluding the Baltic States; aus: Australia; can: Canada; xla: South America, excluding Brazil and Peru; usa: United States. As indicated in Kløverpris (2008), this aggregation covers 92% of the total net expansion.
[e] The maximal and minimal range are based on the qualitative description of the uncertainty related to the biomes conversion results made by Kløverpris (2008). The levels identified as "very good", "good" and "moderate" were considered as an uncertainty of ±20%, 40% and 60%, respectively.
[f] Searchinger et al. (2008). The original values of Searchinger et al. (2008) were however presented for 30 years. The values presented here are converted for 20 years of uptake.

4.3.5 Reacting crops

The environmental impacts related to the cultivation occurring on the newly converted lands have only been considered in one of the case studies performed (Hamelin *et al.* ***V***). The procedure used in Hamelin *et al.* (***V***) to this end is summarized herein. This procedure comports two main steps:

(i) Determine the crop cultivated in each of the converted biomes;
(ii) Estimate the environmental impacts related to the cultivation of the crops identified in (i), for each biome converted.

As non-cultivated lands are converted to agriculture as a result of displaced barley or maize from Denmark, it can be expected that another carbohydrate crop (e.g. wheat, maize, rice, barley, sorghum, millet, rye and oats), or a mixture of such carbohydrate crops, will be cultivated on these lands, in response to the induced price increase on the carbohydrate crop market. This rationale was used in Hamelin *et al.* (***V***), where the reacting crops were considered to be carbohydrate crops only (i.e. a crop among wheat, maize, rice, barley, sorghum, millet, rye and oat). For each of the main regions where expansion takes place (Table 12, Table 15), one reacting crop has been identified (Table 16). This has been done based on data from FAOSTAT (2012e) on the quantity produced of these crops (in each region), for the years 2001-2010. For each region, there was typically one of the carbohydrate crops that have been much more produced than the others over this time period. This crop was then designated as the "reacting crop". When two crops had close production volumes, the one with the greatest increase was considered as the "reacting crop". The results of this procedure are presented in Table 16.

Table 16. Identification of the reacting crops to be cultivated on the converted land

Region	Sub-saharan Africa excluding SACU[d] (xss)	EU-15, excluding Denmark (xeu15)	Brazil (bra)	Former Soviet Union, excluding the Baltic States (xsu)	Australia (aus)	Canada (can)	South America, excluding Brazil and Peru (xla)	United States of America (usa)	Rest of the world	Total
Net expansion[a] (m^2)	367	361	217	182	144	107	87	68	125	1658
Reacting crop[b]	Maize	Wheat	Maize	Wheat	Wheat	Barley	Maize	Wheat	-	
Country[b]	Botswana	France	Brazil	Kazakhstan	Australia	Canada	Argentina	U.S.	-	
Crop share in the "reacting crop[c]" mix (%)	24	24	14	12	9	7	6	4	-	100

[a] Based on Kløverpris (2008), i.e. these are the figures used to derive the figures appearing in column (A) of Table 12. The total appearing in Table 12 does not include the share from "the rest of the world", and is given with only 2 significant digits. As a result, it differs from the total presented here.
[b] Defined based on FAOSTAT (2012e), as described in the text.
[c] The share is calculated excluding the land conversion occurring in "the rest of the world".
[d] SACU: South African Customs Union: Botswana, Lesotho, Namibia, South Africa, Swaziland.

The life cycle data used to estimate the environmental impacts related to the cultivation of all reacting crops identified in Table 16 (step ii) were based on the Ecoinvent v. 2.2 database (Frischknecht & Rebitzer, 2005), and are further detailed in the SI of Hamelin *et al.* (***V***) (Table S18).

5. CARBON FLUXES IN BIOSYSTEM LCAS

5.1 Is biogenic C "neutral"?

Many bioenergy LCAs (Carpentieri *et al.* 2005; Styles & Jones 2007; Huo *et al.* 2009; Hillier *et al.* 2009; Brandão *et al.* 2011), or even policies (e.g. European Union 2003, 2009) omit to account for the CO_2 released when biomass is burned, endorsing the assumption that biomass combustion, regardless of the biomass source, would not result in C accumulation in the atmosphere.

In the light of section 4, it is clear that biomass burning does not necessarily imply that there is no net C accumulation in the atmosphere. In fact, bioenergy systems also need to provide feed/food (and this in a context where land is a finite resource), just like the fossil fuels system does. To the extent that energy crops end up displacing natural ecosystems, the carbon released from soils and vegetation, plus the lost future sequestration, does generate additional C in the atmosphere.

It may however be true that burning biomass results in less C in the atmosphere if:

i. The land used for energy crops would have otherwise grown no plants (e.g. irrigation of desert, or any land where no natural vegetation could be established by itself);
ii. The food/feed crops displaced by energy crops are not replaced;
iii. The food/feed crops displaced are replaced fully by intensification[43];
iv. The biomass used for bioenergy would have otherwise been left to decay, and that biomass would not have contributed to long-term C sequestration into soil (e.g. roadside grass).

The common denominator of all these cases is that no food/feed crop is displaced (or if displaced, not replaced) and no C sequestration capacity is lost. In this sense, they all provide (except case ii) what recent publications refer to as "additional C" (Searchinger *et al.* 2009; Searchinger 2010; Smith & Searchinger 2012; Haberl *et al.* 2012), i.e. C that would otherwise be in the atmosphere if not incorporated in biomass used for fuel.

However, while case (i) appears little realistic to supply a large-scale bioenergy demand like the one created by prominent legislations and targets worldwide (GBEP/FAO 2008), case (ii) involves hunger and starvation and is, thus, not a desirable way to reduce GHG. Although case (iii) is equally not realistic (e.g. Tilman 1999; Kløverpris 2008; Marelli *et al.* 2011; Foley *et al.* 2011) it must be acknowledged that any biomass obtained through intensification does represent an additional biomass, i.e. a feedstock grown in excess of that needed in a situation without bioenergy (this is also true for case i). Such biomass is, however, not GHG-neutral either, as it likely requires additional input like e.g. N fertilizers, which often results in additional GHG emissions (section 10.4.4). Case (iv) is realistic up to a certain scale (for example, the global residues potential is estimated to ca. 100 EJ y^{-1}: Smeets *et al.* 2007; Dornburg *et al.* 2010; Haberl *et al.* 2010), and is typically well acknowledged as the most sustainable bioenergy feedstock (European Commission 2010; Havlík *et al.* 2011). However, if a part of the residues would have otherwise contributed to permanent C sequestration into soil, as this is often the case with e.g. straw incorporated to agricultural soils, bioenergy then involves an increase in atmospheric CO_2 in comparison to the situation without bioenergy (at least over the time period where the C stays in the soil).

It may be argued that to the extent indirect land use changes and foregone sequestration are included and that a proper system expansion is performed, there is no need to take biogenic C from the bioenergy

[43] This of course depends on the type of intensification that is performed. If this generates an increase of fossil fuels use (e.g. for machinery, for producing additional agro-chemicals, etc.), then it may not hold true. Further, even though intensification could result to less C in the atmosphere, the overall amount of GHG in the atmosphere could still be increased (e.g. if N_2O emissions are drastically increased).

feedstock into account, on the basis that no net CO_2 is emitted (the biomass burning only returning the C absorbed by growing plants back to the atmosphere). In many cases, this may however be mathematically wrong for 3 main reasons:

i. Part of the C absorbed by the biomass may not be returned to the atmosphere, if C sequestration into the permanent soil C pool occurs, for a given crop system (in this case, tailpipe emissions are lower than the uptake, involving a net reduction in atmospheric C. This was for example the case in most of the perennial crop systems as well as some annual crop systems studied by Hamelin *et al.* ***I***). Conversely, some crop systems may result into the loss of native soil C (Hamelin *et al.* ***I***).

ii. Part of the C absorbed by biomass may be emitted not as CO_2, but as CH_4 (if the biomass, or part of the biomass, is exposed to anaerobic conditions, whether intentionally or not), in which case a greenhouse gas with a global warming potential 25 times the one of CO_2 (over a 100 y horizon) is emitted (IPCC 2007)[44].

iii. The CO_2 emissions from temporary C losses, before being captured by biomass regrowth, do spend some time in the atmosphere, during which they do contribute to radiative forcing, and thus to global warming (Cherubini *et al.* 2011).

In a nutshell, both fossil fuels and biomass burning result in a net transfer of C to the atmosphere, and atmospheric reactions do not distinguish between fossil and biogenic C. When fossil fuels are burned, C that was stored underground is transferred to the atmosphere, while when energy crops are burned, C that would have otherwise been stored by plants and soils is transferred to (ILUC) or remains in (foregone sequestration) the atmosphere. Whether bioenergy results in less atmospheric C or not depends on which effect outweighs the other (Haberl *et al.* 2012).

In this PhD work, the C flows (both as emissions and removals) involved in the different bioenergy scenarios considered are fully accounted for at each stage of the life cycle via system expansion, state-of-the-art mass balances and dynamic modeling (the latter applying for soil C changes only, as described in section 6). This way, the overall change in atmospheric CO_2 induced by bioenergy systems (as compared to the reference case where bioenergy does not occur) is transparently accounted for.

Detailed analyses on the implications of omitting to account for biomass tailpipe C- emissions in policies can be found in Searchinger *et al.* (2009), EEA Scientific Committee (2011) as well as in Bird *et al.* (2012), among others. Similarly, explanations on the origin of the practice to consider biogenic CO_2 as neutral, which would be a mere misinterpretation of the original guidance provided for the national level carbon accounting under the United Nations Framework Convention on Climate Change (UNFCCC), are provided in Haberl *et al.* (2012).

5.2 Time horizon

In most LCA studies, the assessment of global warming is based on the IPCC AR4 methodology (IPCC 2007). This methodology uses an indicator, namely cumulative radiating forcing (CRF), to derive global warming potentials (GWP) for different substances, which can be used to sum up the different GHG inventoried. GWPs are calculated as the ratio between the CRF of a non-CO_2 GHG and the CRF of CO_2, both integrated over a given time horizon (TH) (the same for both) (IPCC, 2007). This time horizon is commonly taken, in LCAs, as 100y (as in the Kyoto Protocol). A TH of 100 years was also used in the case studies performed within this PhD work.

[44] Although this CH_4 portion is typically accounted for, the C balance becomes mathematically incorrect if the uptake and tailpipe CO_2 are ignored, but the CH_4 accounted for. In fact, the sum of C outputs (here as CO_2 and CH_4 emissions) should be equal to the sum of C inputs (here C uptake). Accounting for the CH_4 only while treating the uptake and tailpipe CO_2 as zero involves that the CH_4 is wrongly treated as an additional release.

The use of this approach may however be seen as a limitation when emission releases occurring at different times (e.g. year 0 and year 13) are involved, as these releases are then summed together despite that their end points of analysis are different (e.g. year 100 and year 113). The IPCC AR4 methodology will in fact treat these releases as occurring at the same time, and doing so involves losing information about the end-point of analysis.

In recent years, a number of studies have proposed methodologies to address this flaw, where many emphasized the particular case of ILUC (e.g. O'Hare *et al.* 2009; Müller-Wenk & Brandão 2010; Kendall 2012). An overview of these methodologies is provided in Brandão *et al.* (2013). As these methodologies are still in their early development stage, the global warming results presented in this study are based on the IPCC methodology. However, the importance of time-dependency was assessed in Tonini *et al.* (***II***) (SI), for the cultivation of *Miscanthus* in Denmark (including ILUC), based on the "time-adjusted warming potentials" described in Kendall (2012)[45]. This specific simulation indicated that accounting for time-dependency would increase the GWP by ca. 40%, and the reasons for this are detailed in Tonini *et al.* (***II***) (SI). Such increase was also suggested by the results of O'Hare *et al.* (2009), for a different bioenergy case.

5.3 Summary of important C flows in energy crop systems

When accounting for C flows in energy crop systems, two main systems may be distinguished:

(i) The plant-soil system: i.e. all processes involved up to crop harvest;
(ii) The post-harvest system: i.e. the system where the harvested crop is used.

A simplified overview of these systems, as needed for performing LCAs, is provided below. It is acknowledged, however, that these systems are in reality much more complex than described below.

In the plant-soil system, C is first absorbed from the atmosphere to the plant (C_{upt}). As plants grow, they produce above- and below-ground residues that will not be harvested ($C_{residues}$), while the remainder of the C absorbed stays in the plant and will be harvested ($C_{harvest}$). Besides C_{upt}, part of the carbon input to the system also origins from the seed (C_{seed}).

Harvestable residues such as straw and beet tops will be part of $C_{residues}$ or $C_{harvest}$, depending on whether they are harvested or not. Depending on the soil C equilibrium where cultivation takes place, part of the C from the non-harvested residues, together with any other eventual C inputs to the soil (e.g. manure) (C_{manure}), will integrate the soil C pool, while the rest will be emitted to the atmosphere ($C_{emiss.cultiv}$), most likely as CO_2, although a small amount may be emitted as CH_4 if there is presence of anaerobic conditions. In some cases, however, the rate of C degradation in the soil may be greater than the C inputs to soil (e.g. residues and manure). When this is the case, there is going to be a loss of soil C to the atmosphere instead of a gain. The difference in soil C as a result of a given cultivation system is referred to as ΔC_{soil}. Other C inputs (C_{other}), like urea or lime, may also be added to the plant-soil system, although these can be considered to be ultimately fully degraded to CO_2 (IPCC 2006a) ($C_{emiss.other}$).

Figure 4 summarizes the main C flows found in the plant-soil system. The main mass balance equations related to this system are presented in Equation 2 and 3:

$$C_{upt} = C_{harvest} + C_{residues} - C_{seed} \qquad \text{(Equation 2)}$$

$$C_{emiss.cultiv} = C_{manure} + C_{residues} - \Delta C_{soil} \qquad \text{(Equation 3)}$$

[45] This methodology has the advantage to be very similar to the IPCC AR4 one. The resulting metric is CO_2 eq. of today. The main difference between this methodology and the IPCC one is the numerator used to calculate the GWP. In Kendall (2012), the bounds of the integral for the CRF of the non-CO_2 substance will be from 0 to "TH minus the year when the emission occurs" (instead of being from 0 to TH, as for the CRF of CO_2). Based on this metric, the more an emission is push into the future, the shorter will be the time horizon over which it is integrated.

Where ΔC_{soil} can be positive (gain in soil C) or negative (loss of native soil C).

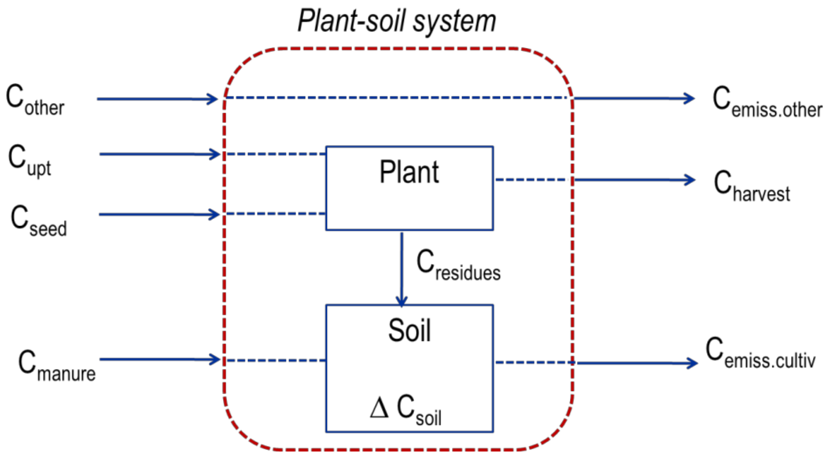

Figure 4. Illustration of the main C flows in the plant-soil system. C inputs are illustrated to the left, while net C output flows are shown to the right.

In the post-harvest system, most of the $C_{harvested}$ is ultimately released again to the atmosphere. This will be essentially through respiration and excreta, if the crop is consumed by humans and animals ($C_{emiss.cons}$). If the crop is used for energy, this will be principally through an exhaust (whether the crop is used as a solid, gaseous or liquid fuel) and through the losses occurring along the biomass-to-energy conversion chain ($C_{emiss.fuel}$). When the crop is used for energy, it can be considered that fossil C (C_{fossil}) is avoided, as the energy would have otherwise been provided by fossil fuels.

The overall C flow in a bioenergy system may be illustrated for the simplified example of an energy crop, say *Miscanthus*, used for CHP. Assuming that barley is the marginal crop displaced, that no manure, lime or urea are applied and neglecting the C inputs from seed, the system may be illustrated as shown in Figure 5. The ILUC system is also included in Figure 5. This implies the C losses from soil and vegetation during land conversion (C_{ALE}), the C involved during the cultivation of the reacting crops (C_{RC}) and intensified crops (C_I) as well as the C from foregone sequestration ($C_{upt.nature}$). This also implies the net C releases from the displacement effect (although not illustrated in Figure 5).

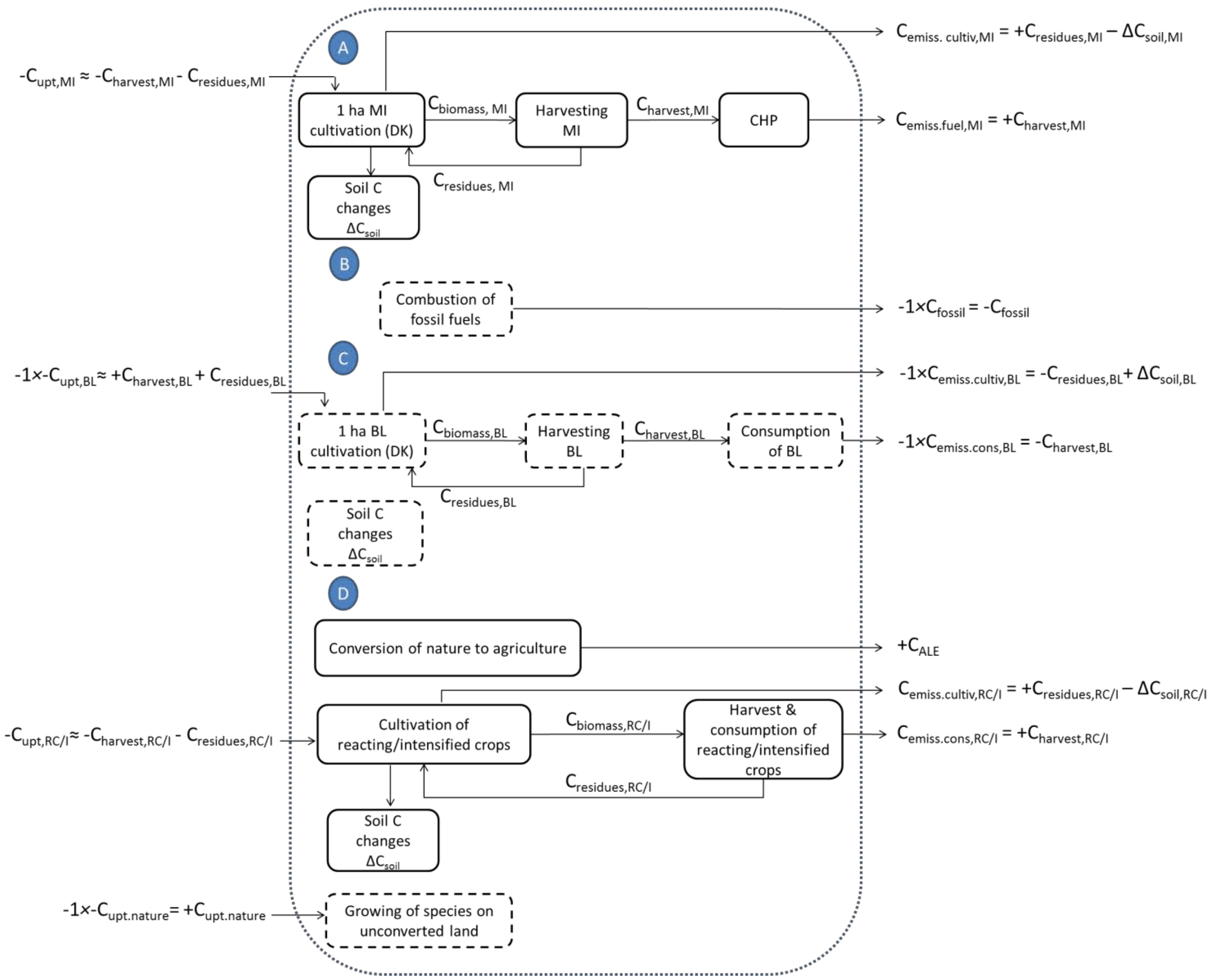

Figure 5. Overview of the main C flows in biosystem LCAs, for a simplified *Miscanthus* to CHP example (see text). Dotted lines indicate avoided C flows: for this reason, these are multiplied by -1. MI: *Miscanthus*; BL: barley. Reacting crop and intensified crop systems are presented together for space constraints, although these are two distinct systems. Flows on the left of the box are input, while flows on the right of the box are the net output. The net C balance represents the sum of all net inputs and outputs.

The net flows from the simplified system illustrated in Figure 5 are the sum of all input flows (left side) and output flows (right side):

- System A: *Miscanthus* cultivation and use for energy
  Net A = $C_{residues,MI} - \Delta C_{soil,MI} + C_{harvest,MI} - C_{harvest,MI} - C_{residues,MI}$
  Net A = $-\Delta C_{soil,MI}$
- System B: Avoided fossil fuel emissions
  Net B = $-C_{fossil}$
- System C: Avoided barley cultivation in Denmark
  Net C = $-C_{residues,BL} - C_{harvest,MBL} + \Delta C_{soil,BL} + C_{harvest,BL} + C_{residues,BL}$
  Net C = $+\Delta C_{soil,BL}$

- System D: Land conversion, cultivation of reacting crops & intensified crops and foregone sequestration
  Net D = $C_{ALE} + C_{residues,RC} + C_{residues,I} - \Delta C_{soil,RC} - \Delta C_{soil,I} + C_{harvest,RC} + C_{harvest,I} - C_{harvest,RC} - C_{harvest,I} - C_{residues,RC} - C_{residues,I} + C_{upt.nature}$
  Net D = $C_{ALE} - \Delta C_{soil,RC} - \Delta C_{soil,I} + C_{upt.nature}$

For space constraints, the displacement effect (due to ILUC; section 4.3.2) could not be illustrated in Figure 5. The C flows from the displacement effect would, however, be modeled as follows, where R is the replaced crops, and D the displaced crops:

- Displacement effect

Displacement effect = Σ(crop replacement – crop displacement)
Displacement effect = $\Sigma(-C_{upt,R} + C_{emiss.cultiv,R} + C_{harvest,R}) - \Sigma(-C_{upt,D} + C_{emiss.cultiv,D})$
Displacement effect = $\Sigma(-C_{harvest,R} - C_{residues,R} + C_{residues,R} - \Delta C_{soil,R} + C_{harvest,R}) - \Sigma(-C_{harvest,D} - C_{residues,D} + C_{residues,D} - \Delta C_{soil,D} + C_{harvest,D})$
Displacement effect = $\Sigma(-\Delta C_{soil,R} + \Delta C_{soil,D})$

The summation sign in the equations above, for the displacement effect, reflects that these crop displacement-replacement effects may occur in several biomes, until land is expanded or intensified. As earlier explained, displacement is in fact an intermediate response only, until final intensification and/or land expansion occurs.

As a result, the net C flows for this system are:

Net C flows = Net A + Net B + Net C + Net D + Net displacement effect

Net C flows = $-\Delta C_{soil,MI} - C_{fossil} - \Delta C_{soil,BL} + C_{ALE} - \Delta C_{soil,RC} - \Delta C_{soil,I} + C_{upt.nature} + \Sigma(-\Delta C_{soil,R} + \Delta C_{soil,D})$

To make this equation generic, MI may be replaced by “energy crop”, and BL by “marginal displaced crop”:

Net C flows biosystem =

$$-\Delta C_{soil,energy.crop} - C_{fossil} - \Delta C_{soil,marginal.displaced.crop} + C_{ALE} - \Delta C_{soil,RC} - \Delta C_{soil,I} + C_{upt.nature} + \Sigma(-\Delta C_{soil,R} + \Delta C_{soil,D})$$

Where:

ALE : agricultural land expansion
RC : reacting crops
I : Intensification
Upt.nature : uptake from “natural” ecosystem, prior to converting it to agriculture (foregone sequestration)
R : replaced crop (from displacement effect)
D : displaced crop (from displacement effect)

Box 1. Net C flows in a simplified bioenergy system (no C from manure, lime or fertilizers, and C from seed neglected), where the energy crop displaces another crop system, which in turns triggers ILUC effects.

This simplified example illustrates that although the system may seems complex, only a very few flows matter for the net balance, as the other flows will eventually cancel out. On a C balance perspective, it is, thus, “correct”[46] to identify these net flows only. In the perspective of estimating the impact of these changed flows on global warming, a full mass balance accounting for all flows is however necessary, as C may be emitted both as CO_2, CH_4, and even CO. Moreover, a complete balance of the N flows is then also needed.

[46] At the light of the simplifications mentioned in the text.

6. ADDRESSING THE DLUC CONSEQUENCES OF BIOENERGY IN DENMARK

6.1 Database

Producing more bioenergy in Denmark has, as earlier discussed, direct land use changes consequences, i.e. the new energy crops will displace a marginal crop, identified as barley (short-term) or maize (long term) (section 2.6). In order to address the DLUC consequences of converting Danish agricultural land from food/feed crops to energy crops, a Danish-specific consequential life cycle dataset was built, which from this point will be referred to as the "DLUC database".

Though LCI databases already exist for some crops (e.g. Ecoinvent), these apply for specific countries that may not fully represent the Danish situation, given that many of the emission flows involved are rather site-specific, and that the management of the crop systems (e.g. fertilizer inputs) differs, among others because of the differences in legislation. Moreover, none of the existing LCI databases, including the Danish-specific LCAFood database, address all the following issues, which are addressed in the present PhD work:

- The partition of biomass between above- and below-ground biomass, and consequently the partition of the C and N contained in the biomass between these different fractions

- Soil C changes: In most LCAs, the C balance is incomplete, the C uptake from the atmosphere being assumed equal to the C harvested plus the C released from decay of plant residues. Yet, these flows are not necessary equal, and a correct balance should take into account the amount of C sequestrated/released from the soil, over the time horizon considered.

- Perennial crops: Though comprehensive LCA inventories do exist for some annual crops (e.g. Nemecek & Kägi 2007; Jungbluth *et al.* 2007), very few, if any, complete LCA datasets are available for perennial crops like *Miscanthus* and willow, albeit LCAs on *Miscanthus* and willow do exist as well as datasets for some grass types.

The structure of the database built within this PhD work is illustrated in Figure 6. The system boundary includes all activities within the cultivation stage (from soil cultivation to harvest) and the reference flow used for each processes is 1 ha of land in a year. A considerable level of details has been included in the inventory, resulting in a total of 528 combinations, including 7 crops, 2 soil types (sandy loam and sand), 2 climate types (wet and dry), 3 initial soil C level (high, average, low), 2 horizon time for soil C changes (20 years and 100 years), 2 residues management practices (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 %). Selected energy crops include 4 annual (spring barley, winter wheat, silage maize, sugar beet) and 3 perennial crops (*Miscanthus*, willow and ryegrass).

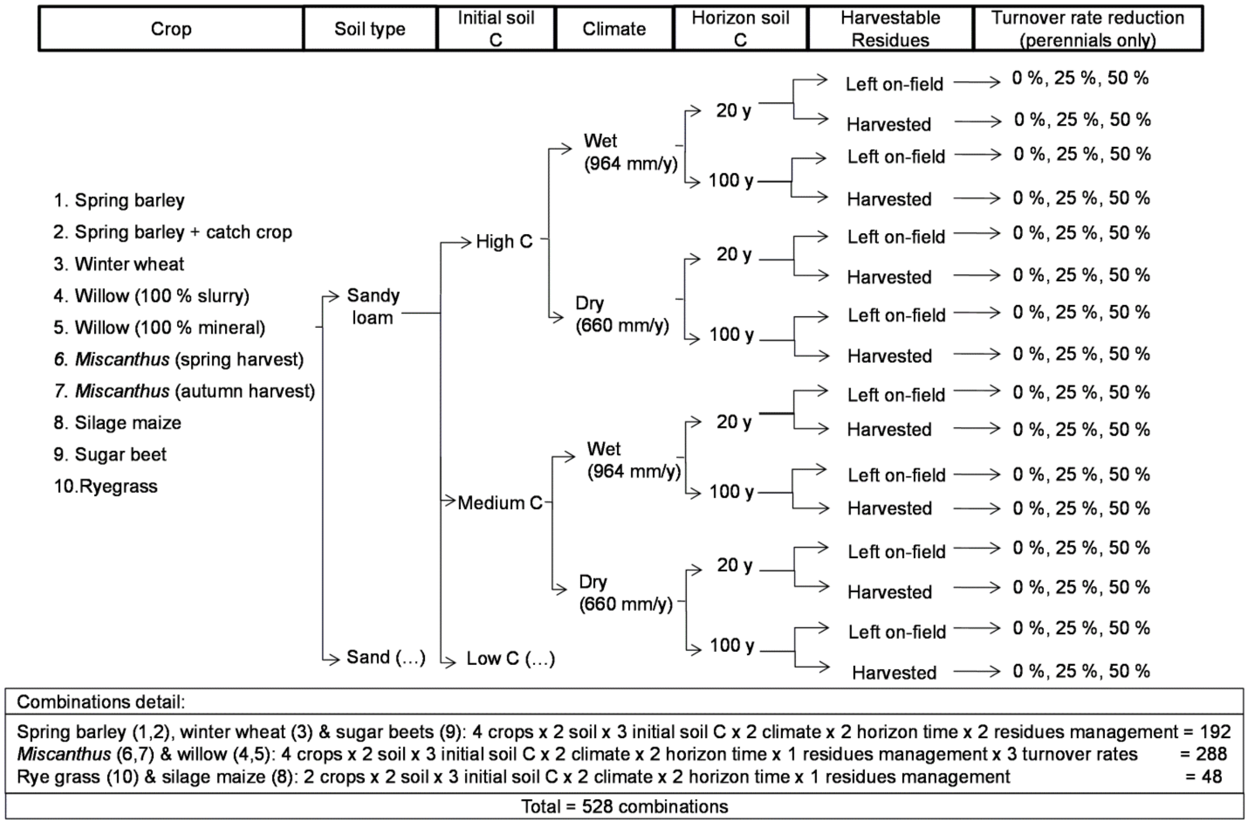

Figure 6. Illustration of the database structure built for addressing the DLUC consequences of bioenergy in Denmark (reproduced with permission from Hamelin *et al.* ***I***)

As shown in Figure 6, the inventory distinguishes between spring and autumn harvest of *Miscanthus*. The reason for this is that the harvest season involves different trade-offs and is likely to influence the conversion route. For example, while the dry matter (DM) yield is higher if harvest occurs in autumn, delaying the harvest to spring involves a lower concentration of minerals in the harvested biomass which favors a better combustion quality (Jorgensen 1997; Lewandowski & Heinz 2003), besides supplying a higher input of C to the soil due to leaves and tops losses occurring during winter. Similarly to *Miscanthus*, spring barley is treated in two different ways (Figure 6) in order to assess the environmental consequences of combining it with a catch crop, which is used for 0.12 to 0.20 Mha of the Danish agricultural area every year. This consists of a non-commercial crop (e.g. perennial ryegrass, oilseed radish) grown to catch the available N in the soil during the autumn period, thereby reducing N leaching (Thorup-Kristensen *et al.* 2003). Though it appears twice in Figure 6, there is only one scenario for willow. This is because willow, as opposed to the other crops, can only be fertilized with either 100% slurry (harvest years) or 100% mineral fertilizers (other years), since slurry spreading is only possible in harvest years with the currently available equipment, under Danish conditions. The life-cycle considered for perennial ryegrass (short-term ley), willow and *Miscanthus* plantations were respectively 2y, 21y (6 cuts; 3 years harvest cycle, but first harvest after 4 years; 1 year establishment; 1 year preparation before planting) and 20y (18 cuts; 1 year establishment: 1 year preparation before planting).

6.2 Life cycle inventory

The database was established within the LCA software SimaPro 7.3.3 (PRé Consultants 2012). Nine main categories of agricultural operations were considered, namely soil preparation, propagation (seed, rhizome or cutting production), liming, sowing/planting, plant protection, fertilization, irrigation, harvest and transport from farm to field. For each of these operations, background data were obtained from the Ecoinvent v. 2.2

database. All modeling details (e.g. specific background processes used for each crop, quantities considered, diesel consumption, etc.) are presented in the SI of Hamelin *et al.* (***I***). Danish regulations (Danish Ministry of Food, Agriculture and Fisheries 2012; Danish Ministry of the Environment 2012) were considered when modeling fertilization, and it was assumed, based on the statistics presented in Nielsen *et al.* (2009), that crops' N demand was fulfilled by 50% animal manure and 50% mineral fertilizers[47]. The modeling of fertilization is further described in Hamelin *et al.* (***I***), in which the fertilization balance for each crop and soil type is also presented. The mineral N, P and K fertilizers considered were calcium ammonium nitrate (CAN), diammonium phosphate (DAP) and potassium chloride, respectively (section 2.5).

6.3 DM, C and N partitioning between harvest, above-ground and below-ground residues

In order to build a life cycle inventory allowing to follow the C and N flows throughout the whole crop continuum, the inventory was structured into four main categories:

- Primary yield: yield of the main product, i.e. the product motivating the cultivation.
- Secondary yield: the harvestable fraction of crop residues (e.g. straw, beet tops).
- Above-ground residues: Non-harvestable crop residues that do not enter the soil (e.g. stubbles, leaves, branches & twigs from woody crops, etc.).
- Below-ground residues: Crop's photosynthetic material entering the soil (e.g. via rhizodeposition, root material, shoot residues, etc.).

The data source and methodology used in order to determine the DM, C and N partition between the four above-mentioned categories, for all *crops×soil×climate* combinations considered in the inventory (Figure 6), are detailed in Hamelin et al. (***I***).

Table 17 presents an overview of the results of this procedure, together with the main C and N inputs considered for all crop systems. For a more visual overview, the partition of the DM is illustrated in Figure 7 (as it can be seen from Table 17, C and N partition results are proportional to those of DM, which is the reason why only DM results are illustrated). From this, it can be seen that *Miscanthus* generates the highest net DM production, while barley generates the lowest. Even though it has a low primary yield (compared to the other crops assessed), wheat still generates a relatively high DM production, due to its high proportion of above- and below-ground residues. Figure 7 also illustrates the trade-offs between harvesting *Miscanthus* in autumn (higher proportion of the DM in the primary yield) or during spring (DM proportion in AG residues greater than the proportion in the primary yield). It further highlights maize as an "efficient crop", for which very little DM ends up in the above- and below-ground residues. One observation that can be highlighted from Table 17 is the relatively lower N inputs required for the *Miscanthus* systems, as well as the important amount of pesticides input needed for sugar beet.

[47] As earlier described, this does not apply for willow, which is fertilized by whether 100% mineral fertilizers, or 100% manure, depending on the years. The manure share was considered to consist of 50% fattening pig slurry, and 50% dairy cow slurry. In calculating the amount of slurry to be applied to crops, an efficiency of 75% was considered for pig slurry, and of 70% for cow slurry, in conformity with the Danish legislation (Danish Ministry of Food, Agriculture and Fisheries 2012).

Table 17. Overview of the main characteristics, inputs and DM, C and N partition for the crops considered in this PhD work, over all *soil×climate* combinations inventoried[a]

		Spring barley	Winter wheat	Willow	Miscanthus[f] (autumn harvest)	Miscanthus[f] (spring harvest)	Maize silage	Sugar beet	Ryegrass
Plant characteristics[b]		Annual, C3	Annual, C3	Perennial, C3	Perennial, C4	Perennial, C4	Annual, C4	Annual, C3	Perennial C3
Plantation duration (y)		1	1	21	20	20	1	1	2
DM, as a % of FM[c]		85%[d]	85%[d]	50%	44%	85 - 90%	31%	22%[d]	18%
Primary yield ($t\ ha^{-1}\ y^{-1}$)	*DM*	4.3 – 4.9	5.6 – 6.8	7.1 – 13	13 – 15	8.5 – 10	12	13	8.8 – 10
	C	1.9 – 2.2	2.5 – 3.1	3.2 – 5.7	5.8 – 6.9	3.8 – 4.5	5.4	5.6	4.0 – 4.5
	N ($\times10^{-3}$)	76 – 87	110–130	40 – 73	65 – 76	17 – 20	170	120	280–320
Secondary yield ($t\ ha^{-1}\ y^{-1}$)	*DM*	2.3–2.7	3.1–3.7	-	-	-	-	1.6	-
	C	1.1–1.2	1.4–1.7	-	-	-	-	0.73	-
	N ($\times10^{-3}$)	2.3–2.7	3.1–3.7	-	-	-	-	1.6	-
AG[e] residues ($t\ ha^{-1}\ y^{-1}$)	*DM*	2.9–3.3	3.8–4.6	5.0–6.0	4.8–5.6	9.3–11	2.1	3.7	4.1
	C	1.3–1.5	1.7–2.1	1.5–2.7	2.2–2.5	4.2–4.9	0.96	1.7	1.9
	N ($\times10^{-3}$)	10	17	17–30	29–34	92–110	6.3	97	26
BG[e] residues ($t\ ha^{-1}\ y^{-1}$)	*DM*	1.9–2.2	4.2–5.0	5.2–6.3	2.8–3.3	2.8–3.3	2.5	2.4	8.3
	C	0.87–0.99	1.9–2.3	1.6–2.8	1.3–1.5	1.3–1.5	1.1	1.1	3.7
	N	27–31	37–45	17–31	14–17	14–17	17–18	34	100
N applied, manure and CAN ($kg\ N\ ha^{-1}\ y^{-1}$)		140–150	190–200	120–170[g]	110	71	170–190	120–150	390–410
P applied, manure and DAP ($kg\ P\ ha^{-1}\ y^{-1}$)		22	22	15–31[g]	15	15	44	43	44–45
K applied, manure and K_2O ($kg\ K\ ha^{-1}\ y^{-1}$)		62–68	87–90	50–130[g]	75	75	135	150	239
C applied with manure ($t\ C\ ha^{-1}\ y^{-1}$)		0.55–0.61	0.80–0.78	0–1.2[g]	0.43	0.29	0.67–0.78	0.49–0.59	1.6–1.7
Lime ($t\ ha^{-1}\ y^{-1}$)[h]		0.17	0.17	0.04	0.04	0.04	0.17	0.17	0.04
Pesticides[i] ($kg\ ha^{-1}\ y^{-1}$)		0.17	2.0	1.4	1.6	1.6	0.80	6.40	0.65

[a] All data are from Hamelin *et al.* ***(I)***. The ranges reflect the differences between soil types (sandy or sandy loam) and climate types (wet or dry). All values are rounded to 2 significant digits.
[b] C3 and C4 indicate the photosynthesis type
[c] DM: Dry matter; FM: Fresh matter
[d] The same value applies for straw, while for beet tops, DM represents 12% of FM
[e] AG: above-ground; BG: below-ground. Values for AG residues do not include the secondary yield (which should be counted as AG residues when straw is incorporated). For spring barley cultivated with a catch crop, a DM input of 1.4 $t\ DM\ ha^{-1}\ y^{-1}$ is considered for the catch crop (total AG and BG residues), which corresponds to a C input of 0.63 $t\ C\ ha^{-1}\ y^{-1}$ and to a N input of $48 \times 10^{-3}\ t\ N\ ha^{-1}\ y^{-1}$.
[f] For established plantation (year 4). Values for the first years of growth (year 2 and 3) are presented in Hamelin *et al.* ***(I)***.
[g] This wide range reflects the difference between the years where fertilization is performed by 100% slurry, and the years where fertilization is performed by 100% mineral fertilizers.

[h] These are annualized value. The amount of lime considered is 0.84 t ha^{-1}, which is applied once every 5 years for annual crops, and once every 20 years for perennial crops.
[i] Sum of all pesticides applied, detail of the actual pesticide products applied are available in Hamelin *et al.* ***(I)***. For perennial crops, values presented here consist of the sum of all pesticides applied annualized over the whole plantation duration.

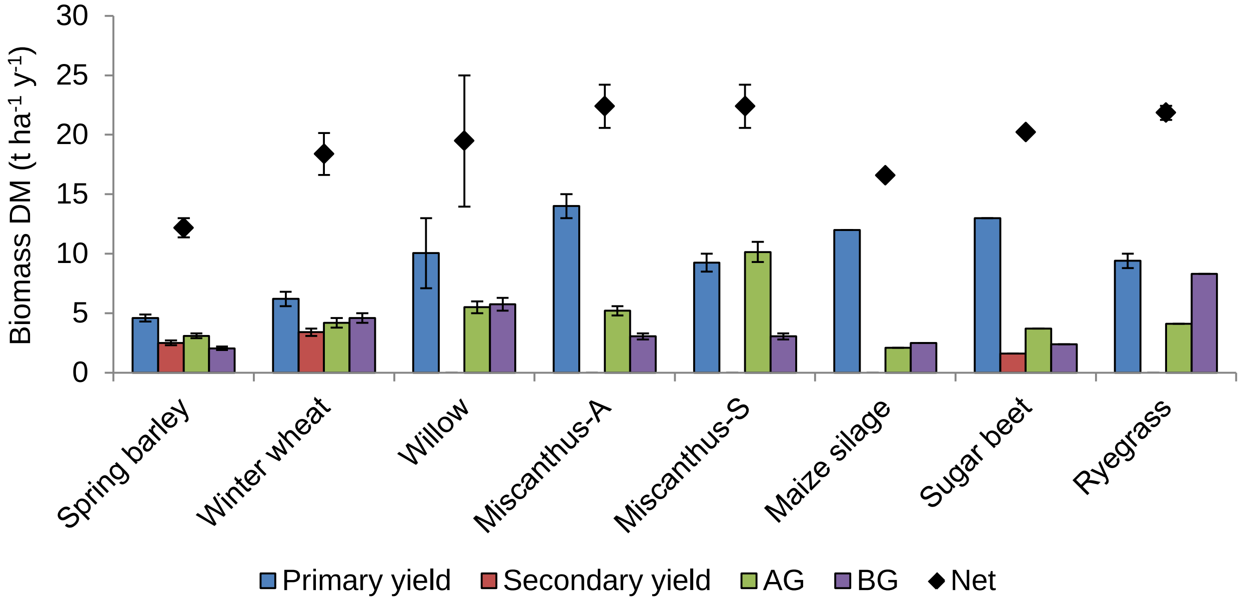

Figure 7. Partition of the DM between primary yield, secondary yield, above- and below-ground residues over all soil and climate combinations considered, based on the values from Table 17. For *Miscanthus*, –A indicates autumn harvest, and –S indicates spring harvest.

6.4 C flows

Soils have an equilibrium C content which is the result of a balance between inflows (e.g. plant matter from above- and below- ground residues, manure, etc.) and outflows (e.g. decomposition, erosion, leaching of soluble C, etc.) to the soil pool. If outflows are greater than inflows, soil C decreases, while soil C increases if inflows are greater than outflows. Output flows are to a great extent determined by climate-specific parameters like temperature and precipitations, where higher temperature and moisture favor the soil biota activity (i.e. decomposition). However, any change affecting the activity of soil biota (e.g. change in oxygen availability due to soil compaction, change in soil pH) will result in greater or smaller decomposition[48]. In this sense, any form of agriculture will disturb the soil equilibrium until a new equilibrium is eventually reached after many years of constant agricultural practices.

Changes in soil C were estimated with the dynamic soil C model C-TOOL, developed to calculate the soil carbon dynamics in relation to the Danish commitments to UNFCCC. This model is parameterized and validated against long-term field experiments conducted in Denmark, UK and Sweden. Further description of the C-TOOL model is given in Petersen *et al.* (2002) and Petersen (2010). Changes in soil C were estimated over two time horizons: 20 years and 100 years. Moreover, an initial "high", "medium" and "low" soil C content were considered (Figure 6). These levels are based on an average of 143.9 ± 59.2 t C ha^{-1} for sandy soils and 144.7 ± 76.4 t C ha^{-1} for sandy loam soils, for the depth 0-100 cm, the medium level being the average, and the high and low levels corresponding to one standard deviation. For *Miscanthus* and willow, the C turnover rate in the topsoil may be reduced in response to the absence of tillage over many years (Olesen *et al.* 2001; Chatskikh *et al.* 2009). In this study, three different turnover rates have been applied for

[48] A decrease of oxygen availability and of the pH will both limit the activity of the soil biota (Cowie *et al.* 2006).

these two crops; no reduction in turnover rate (as for other crops), 25% reduced turnover rate and 50% reduced turnover rate.

The portion of the C input to the soil (i.e. from manure, straw/tops and non-harvestable residues) that does not enter the soil C pool over the time horizon considered is assumed to be lost as a CO_2-C emission to the atmosphere. Similarly, all losses of native soil C are assumed to be transferred to the atmosphere as CO_2-C.

Each mole of lime applied to soil has the net potential to contribute to the addition of 1 mole of CO_2 to the atmosphere, based on the following equation (EMEP/EEA 2007):

$$CaCO_3 + CO_2 + H_2O \rightarrow Ca^{2+} + 2HCO_3^- \rightarrow Ca^{2+} + 2OH^- + 2CO_2 \qquad \text{(Equation 4)}$$

The net result of applying 1 mole of $CaCO_3$ can thus be simplified as:

$$CaCO_3 \rightarrow CaO + CO_2 \qquad \text{(Equation 5)}$$

Considering the molecular weights (100.086 kg for $CaCO_3$ and 44.009 kg for CO_2), it can be considered that applying 1 kg of $CaCO_3$ contributes to 0.44 kg of CO_2 (44.009 kg / 100.086 kg), or 0.12 kg CO_2-C. This estimation implies the assumption that all the lime applied reacts, which may not necessarily be the case in practice. Emissions of CO_2 from application of $CaCO_3$ to the field are estimated by similar rationale in the IPCC guidelines (IPCC 2006c) as well as in (EMEP/EEA 2007).

Conformingly to IPCC (2006a), methane emissions in agricultural fields are typically assumed as negligible, due to the aerobic conditions found in cultivated mineral soils. This assumption was also made in this inventory, where the oxidation of plant matter and manure was only considered in terms of CO_2.

Mineral soils, especially when unmanaged, are often considered to be a sink for atmospheric CH_4, as a result of the microbial oxidation of methane to CO_2 occurring in aerobic soils (Smith et al. 2000; Hutsch 2011)[49]. This sink, however, is relatively small for soils under agricultural cultivation (Smith et al. 2000, Hutsch 2011). Based on measurements results (e.g. Boeckx *et al.* 1997; Robertson *et al.* 2000; Smith *et al.* 2000; Hutsch 2001), not only are these flows very minor cropland systems[50] (this is however different for unmanaged soils), but they were also found to be of similar magnitude among different crop systems involving both annual and perennial crops (Robertson *et al.* 2000; Drewer *et al.* 2012). Although some LCAs (e.g. Brandão *et al.* 2011) incorporated this effect as a function of the amount of N applied (since soil CH_4 uptake is influenced by fertilization), very limited information is available to establish such a link, and the uptake response to fertilization is generally negligible for manure and nitrate-based fertilizers (Hutsch 2001). For these reasons, it has not been included in this study, when dealing with direct land use changes.

Figure 8 illustrates a typical output from C-TOOL, for the case of barley and spring harvested *Miscanthus*, cultivated on a sandy soil and under a wet climate, and considering a 25% reduced C turnover rate due to the absence of tillage over many years (*Miscanthus* only). It shows how an equilibrium is gradually establishing as the crop is cultivated, year after year, on a given field. The results are here presented for 200 years, which is of course not realistic: a given hectare of land will obviously not, in practice, be cultivated with barley or *Miscanthus* during 200 years. However, it illustrates how the change in soil C is much more pronounced at the beginning of the period, before to finally level out and reach equilibrium.

[49] On the contrary, organic soils are often net CH_4 emitters.

[50] The C flows breakdown illustrated in Figures S13-S15 of Tonini *et al.* (***II***) show lime to represent the smallest C flow, with 0.005 t C ha^{-1} y^{-1}. Yet, the average CH_4 uptake from agricultural soils represents, based on available published data, ca. 0.0006 t C ha^{-1} y^{-1} (Robertson *et al.* 2000; Boeckx *et al.* 1997; Hutsch 2001). Based on this, this flow has been considered negligible within this PhD work.

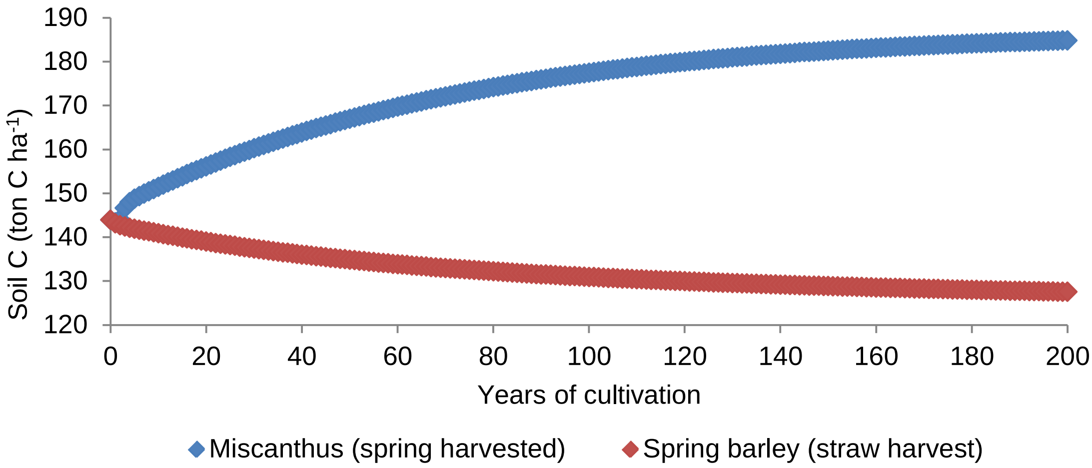

Figure 8. Soil C changes and establishment of an equilibrium after 200 years of cultivation of spring barley (with straw harvest) or spring harvested *Miscanthus*. The cultivation takes place on a sandy soil and under a wet climate. A 25% reduced C turnover rate due to the absence of tillage over the crop 20 y life cycle is considered for *Miscanthus*.

The differences in soil C changes for all assessed crop systems are shown in Figure 9 (reproduced from Hamelin *et al.* ***I***), for 20 years of cultivation on both sand and sandy loam soils. A positive value indicates a gain in soil C, while a negative value indicates a loss of soil native C.

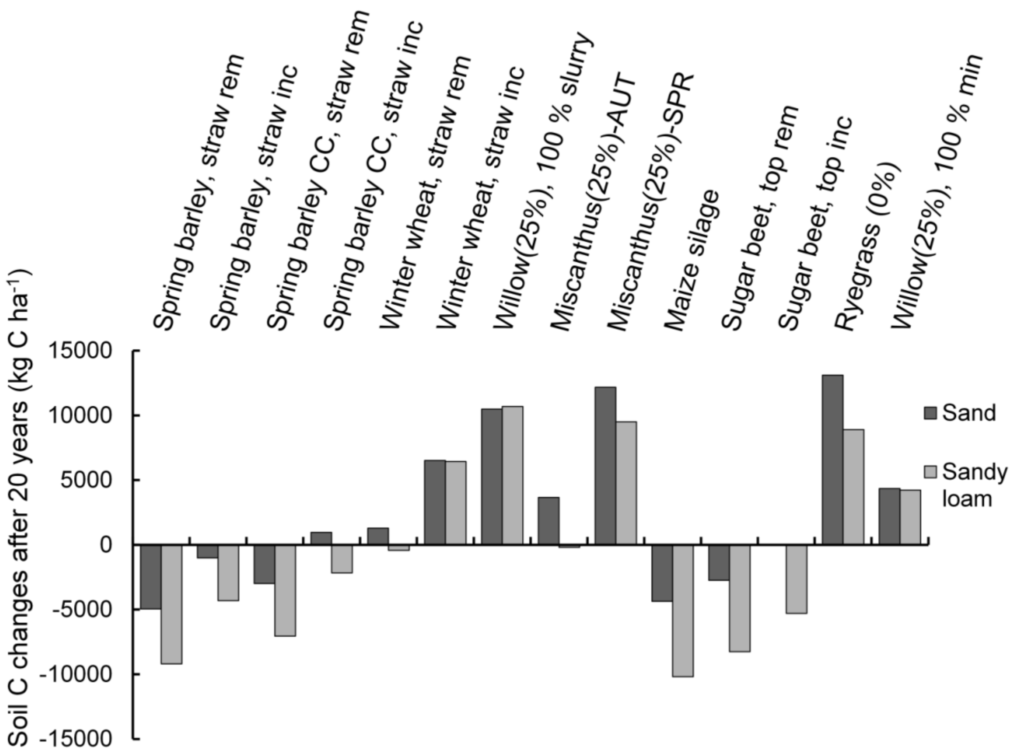

Figure 9. Soil C changes after 20 years of cultivation (reproduced from Hamelin *et al.* ***I***, with permission). Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values between parenthesis for *Miscanthus* and willow indicate the turnover rate reduction considered because of the absence of tillage. Results are for a medium initial soil C and a wet climate.

Additional results, like the influence of wet and dry climates, of the turnover rate reduction for perennial crops and of the initial soil C level on the modeled soil C changes are available in Hamelin *et al.* ***(I)***.

Based on the soil C changes results, on the data presented in Hamelin et al. ***(I)*** (and summarized in Table 17) and on Equations 2 and 3, the full C balance for all crop systems could be established. An example is presented in Table 18 for cultivation on sandy soils, under a wet climate. Results for other *soil×climate* combinations are presented in the SI of Hamelin et al. ***(I)***.

Table 18. Carbon balance for all crop systems on sandy soil, for a wet climate. All values in kg C ha^{-1} y^{-1}. Eventual inconsistencies are due to rounding. (I) indicates incorporation of the secondary yield, and (R) indicates removal (or harvest) of the secondary yield.

Crop systems	Spring barley (R)	Spring barley (I)	Spring barley & catch crop (R)	Spring barley & catch crop (I)	Winter wheat (R)	Winter wheat (I)	Willow (100 % slurry)	*Miscanthus* (autumn) Year 4-20	*Miscanthus* (spring) Year 4-20	Maize silage	Sugar beet (R)	Sugar beet (I)	Rye-grass	Willow (100 % mineral)
INPUTS														
C uptake	5087	5087	5716	5716	7402	7402	8926	10359	10359	7497	9094	9094	10082	8926
C seed[a]	33	33	34	34	78	78	450	540	540	8	0.8	0.8	9	450
C manure	608	608	526	526	801	801	1158	434	289	782	593	593	1650	0
C lime	20	20	20	20	20	20	5	5	5	20	20	20	5	5
OUTPUTS														
CO_2-C, manure and residues	2764	3816	3312	4316	4303	5431	5240	4288	6080	2864	3358	4086	6595	4388
CO_2-C, lime	20	20	20	20	20	20	5	5	5	20	20	20	5	5
C primary yield	1913	1913	1913	1913	2525	2525	4770	6863	4500	5423	5603	5603	4491	4770
C secondary yield (when harvested)	1052	0	1052	0	1388	0	0	0	0	0	728	0	0	0
CO_2-C, from native soil C losses	248	51	149	0	0	0	0	0	0	218	138	0.5	0	0
Δ SOIL C, ANNUALIZED OVER 20 YEARS[b]														
Accumulation/losses of soil C[c]	-248	-51	-149	48	65	326	524	183	609	-218	-138	-0.5	656	218

[a] Data considered for the C contribution of the seed are detailed in the SI of Hamelin et al. ***(I)***
[b] The overall soil C change occurring over a period of 20 years is assumed to be equally (linearly) distributed over all years.
[c] A negative value indicates losses of native soil C, whereas a positive value indicates soil C gains.

6.5 N flows

In the cropping systems considered in this study, there are three main inputs of N: from fertilizers, from crop residues, and from the atmosphere. The output flows considered are ammonia (NH_3), nitrous oxide (N_2O) (direct and indirect), nitrogen oxides (NO_x), emissions of dinitrogen (N_2) to the atmosphere and nitrate (NO_3^-) leaching to ground- and surface waters.

Two NH_3 flows were estimated: the NH_3 from the application of mineral fertilizers and the NH_3 from the application of animal slurry. The emission factors considered for estimating the NH_3 emissions from animal slurry and mineral fertilizer applications (distinguished per fertilizer type) are presented in Hamelin *et al.* (***I***). Other sources could have been considered in estimating NH_3 flows, namely the decaying crops residues as well as the NH_3 emission from crop foliage. Crop foliage was not included as a source of NH_3 emission due to the contradicting results and evidences found regarding the quantification of this emission and its actual occurrence in arable cropland. Similarly, the NH_3 emission occurring as a result of the decomposition of crop residues was not included, because this emission is practically insignificant when residues are incorporated (De Ruijter *et al.* 2010), which is the case for the annual crop systems in this study. For perennials, it is considered that when crops shed their leaves, these are already emptied of easily convertible N (primarily at the profit of storage organs) which should also result in negligible NH_3 emissions from the residues.

Nitrogen oxides consist of the sum of nitric oxide (NO) and nitrogen dioxide (NO_2). Once emitted from the soil (mostly as a result of nitrification), NO is quickly oxidized to NO_2 by available oxidants in the atmosphere (typically ozone) (Delon *et al.* 2008). Though gaseous NO_2 is emitted from biological processes occurring in the soil (Graham *et al.* 1997), no information has been found on NO_2 emissions from soils in the selected crop systems. Therefore, the emissions of NO are assumed to represent total NO_x. Stehfest & Bouwman (2006) reported NO-N emissions for Europe of 144 Gg y^{-1}, for a N application of 12812 Gg y^{-1}, for cropland. Based on this, an emission factor of 0.011 kg NO-N per kg N applied can be derived. Similarly, an emission factor of 0.013 kg NO-N per kg N applied was derived for grassland. For crop residues, based on Haenel *et al.* (2010), an emission factor of 0.007 kg NO-N per kg N was used.

The formation of N_2O in crop systems is particularly favored by partial or transient anaerobic conditions, but also by high concentrations of NO_3^- in the soil solution, by the presence of an available carbon source, and by warm temperatures, among others (Nieder & Benbi 2008; Stehfest & Bouwman 2006). Further, Thomsen *et al.* (2010) proposed a model suggesting that the ratio of the oxygen demand to the oxygen supply of a particular field system has significant influence in determining whether emissions of N_2 (complete denitrification) or N_2O will be favored. This is further elaborated in section 10.4.3. Because of this dependence upon such site-specific factors, emissions of N_2O exhibit a very high degree of spatial and temporal variability. Emissions of N_2O from cultivation activities are, for LCI, generally estimated based on extrapolation from field measurements, from biogeochemical models or most commonly calculated based on IPCC guidelines (IPCC 2006c). Based on this methodology, N_2O emissions are assumed to be proportional to the N content of the source material for N_2O emissions. Though critiques have been published about the IPCC methodology (e.g. Jungkunst & Freibauer 2005; Smeets *et al.* 2009; Stehfest & Bouwman 2006), the use of IPCC default factors for estimating N_2O emissions is probably the best methodology to use outside well-characterized areas (Edwards *et al.* 2008). In Denmark, IPCC default emission factors approximately correspond with measured emissions (e.g. Chirinda *et al.* 2010a), and this finding has also been reported for other trials around Northern Europe (e.g. Hellebrand *et al.* 2010; Schouten *et al.* 2012; Don *et al.* 2012). For these reasons, it is the IPCC methodology (IPCC 2006c) that is applied in this study to estimate the N_2O emissions from the different crop systems. Based on this methodology, an emission factor of 0.01 kg N_2O-N per kg N for synthetic fertilizer, organic fertilizer and crop residue inputs is considered. A portion of the volatilized N (as NH_3 and NO_x) that is re-deposited will subsequently be emitted as N_2O. Similarly, a portion of the N losses through leaching may be emitted as N_2O. These are referred to as indirect N_2O emissions (IPCC 2006c). The IPCC methodology suggests an emission factor of 0.010 kg N_2O-N per kg NH_3-N plus NO_X-N volatilized and of 0.0075 kg N_2O-N per kg N leaching. The IPCC methodology also suggests to account for N_2O emissions occurring as a result of soil organic matter mineralization, in situations where native soil C is lost due to a change in land use or management. This has not been considered in this inventory, because of the inconsistency in the IPCC (2006b) approach that only considers emissions of N_2O from losses of soil organic matter and not reductions in estimated N_2O emissions when soil organic matter is accumulated. In fact, the better soil structure and soil aeration associated with higher soil organic matter levels was observed to lead to reduced N_2O emissions (Chirinda *et al.* 2010b)[51]. However, this effect was included as a sensitivity analysis, in order to size the importance of this contribution to the overall N_2O emissions.

Leaching of N was, for ryegrass and annual crops, calculated with the N-LES_4 model (Kristensen *et al.* 2008), a continuously updated empirical model to predict N leaching from arable land based on more than 1200 leaching studies performed in Denmark during the last 15 years. For *Miscanthus*, N leaching estimates were based on data from Olesen *et al.* (2001), and these estimates are further detailed in the SI of Hamelin et al. ***(I)***. Nitrate leaching for willow was considered to be the same as for *Miscanthus*. For both *Miscanthus* and willow, N leaching has been considered to be highest in the planting year (Mortensen *et al.* 1998).

6.6 P flows

Phosphorus losses from agricultural soils have been estimated as 5% of the net surplus application, based on Nielsen & Wenzel (2007). For perennial crops, P losses were estimated as 2.5% of the net surplus application, based on Sørensen *et al.* (2010). This reflects their lower risk for erosion in comparison to annual crops (Börjesson 1999).

6.7 Other flows

Biogenic non-methane volatile organic compounds (NMVOC) emitted from photosynthesizing leaves of crops (particularly isoprene and monoterpene) were taken into account in the inventory. The calculation of NMVOC in this study is based on the methodology described in Haenel *et al.* (2010) considering specific emission factors (in kg NMVOC DM^{-1} h^{-1}) for the different crop systems, as detailed in the SI of Hamelin *et al.* ***(I)***.

Copper (Cu) and zinc (Zn) losses, which are of relevance for toxicity-related environmental impacts, were estimated similarly as for P losses, though it was assumed that 100% of the surpluses were lost (Wesnæs *et al.* 2009). The calculation of P, Cu and Zn losses is detailed in the SI of Hamelin *et al.* ***(I)***.

Though the importance of understanding the fate of K and to some extent of calcium (Ca) on an agronomical perspective is recognized, K and Ca losses towards soils and waters are not flows affecting any of the environmental impacts categories described in the Danish EDIP method for life cycle impact assessment, which was, as earlier mentioned, used throughout this PhD work. The fate of K and Ca was therefore not considered further.

6.8 Sensitivity analyses

In the inventory results presented in Hamelin et al. ***(I)***, the N requirements of crops were fulfilled with 50% manure and 50% mineral fertilizers, here considered to be CAN. Though this reflects well the conditions of Denmark, a world-leading exporter of pig and dairy products where considerable volumes of manure are available (Dalgaard *et al.* 2011), it does not represent the situation of countries where manure access is limited. Because of the manure consequences on the C and N balances of the crop systems, a sensitivity analysis was carried out to reflect the situation where fertilization is provided by mineral fertilizers only. As shown in Hamelin *et al.* ***(I)***, the impact of this translates mostly in terms of soil C changes, highlighting the

[51] On the other hand, it can be argued that an increased soil organic matter input to soil results to net increases of N_2O emissions, due to both the higher N input and the increase of microbial activity stemming from the higher C content (e.g. Li *et al.* 2005). However, based on the measurement results presented in Stehfest & Bouwman (2006), it is here considered that for soils with soil organic C content below 3%, like those typically cultivated in Denmark, the dominating effect is most likely a net N_2O reduction due to the improved soil structure.

importance of manure inputs for reducing soil C losses (or enhancing soil C gains). This is shown in Figure 10a. Figure 10b shows, on the other hand, the differences in NH_3 emissions if the mineral fertilizer portion is provided by urea (the marginal N fertilizer on a world perspective; section 2.5) instead of CAN. Besides NH_3, this also slightly affected N_2O through the indirect N_2O flows due to volatilization (i.e. the N_2O from the portion of re-deposited ammonia and NO_x), and induced an additional CO_2 release due to the degradation of urea. As these were shown to be minor (Hamelin *et al.* ***I***), only the changes of NH_3 flows are illustrated here.

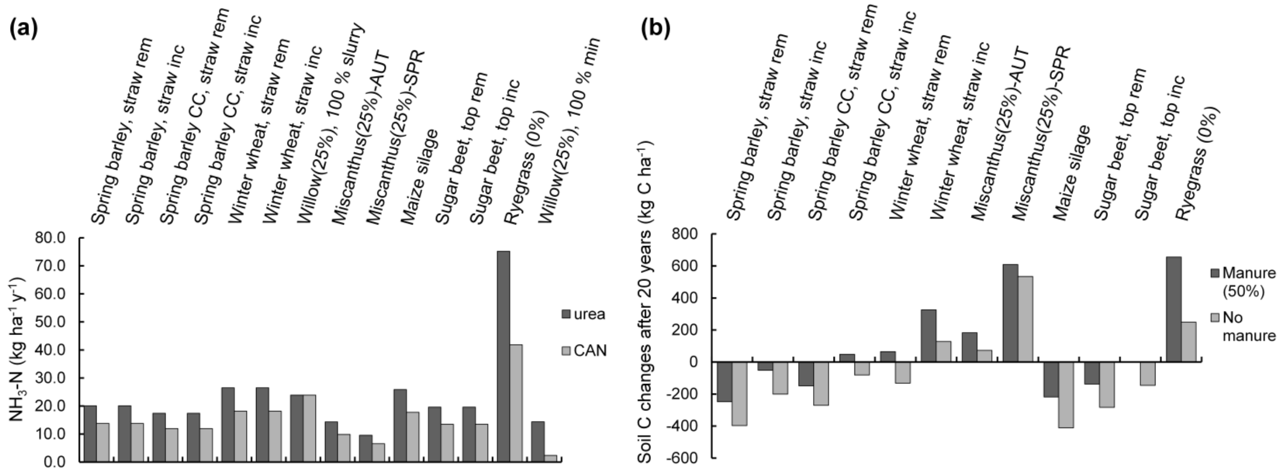

Figure 10. Sensitivity analyses for: type of N mineral fertilizer used versus NH_3 emissions (a) and use of manure versus soil C changes (b). Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values between parentheses for perennial crops indicate the turnover rate reduction considered because of the absence of tillage. Results are for a sandy soil with a medium initial soil C and under a wet climate.

As further detailed in Hamelin et al. ***(I)***, sensitivity analyses were also carried out in relation to the calculation of N_2O, where two different methodologies were used:

i. IPCC methodology (with and without the inclusion of additional N_2O due to the mineralization of soil organic matter)
ii. Top-down approach described in Crutzen *et al.* (2008) (using their 3% and 5% emission factors)

The results are presented in Figure 11.

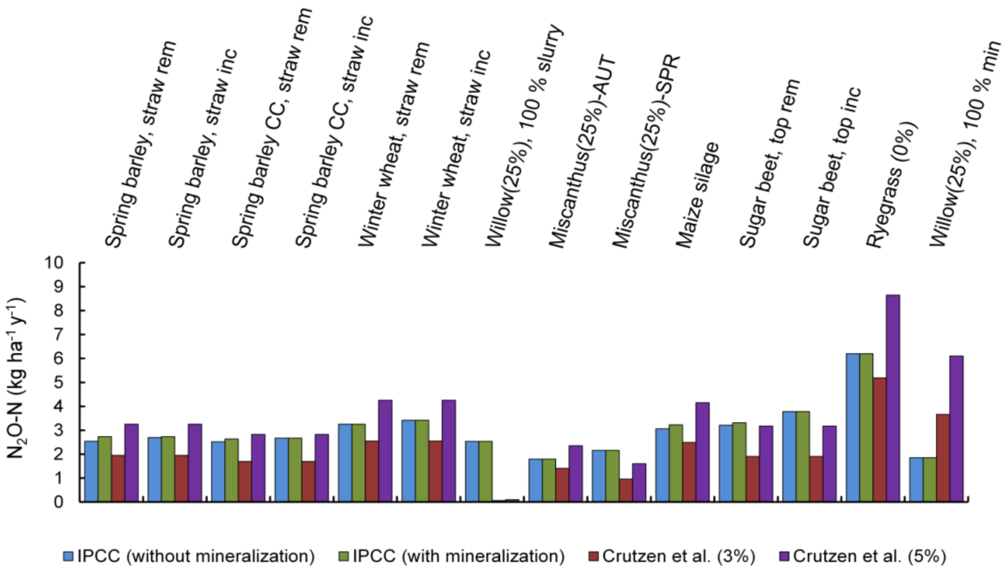

Figure 11. Sensitivity analysis results for the type of methodology used to calculate N_2O emissions. Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values between parentheses for perennial crops indicate the turnover rate reduction considered because of the absence of tillage. Results are for a sandy soil with a medium initial soil C and under a wet climate.

6.9 Characterized inventory results per crop and comparison with other database

The objective of the study performed in Hamelin *et al.* (***I***) was to build a state-of-the-art LCA database allowing to include the direct land use change consequences of bioenergy production in Denmark. In Figure 12, the characterized inventory results per crop are presented[52], for the 4 main impact categories considered in this PhD work (section 2.1). These, however, should be seen as “cradle-to-gate” results rather than results reflecting the full crop life cycle. In fact, the inventory ends after crops’ harvest: it does not include the storage, neither the final fate of the crop (e.g. combustion for energy production, consumption for calories intake, etc.). This applies especially for the global warming results (Figure 12a), where most of the C stored in crops and straw/tops will ultimately be released as CO_2, but still appears as a sequestration in the balance when the “gate” is at the harvest stage. For example, it is for this reason that crops involving harvest of the secondary yield (i.e. straw/top removal) feature, in Figure 12a, a lower global warming potential than crops for which the secondary harvest is incorporated. Further, it should be highlighted that the results presented in Figure 12 are simply the inventory results and not a LCA per se. For example, these results do not include, as a full LCA would, the avoided production and use of mineral fertilizers related to the portion of manure used to fulfill crops N, P and K needs.

52 For perennial crops, these represent the full crop life cycle inventory, annualized over 20, 21 and 2 years for *Miscanthus*, willow and ryegrass, respectively.

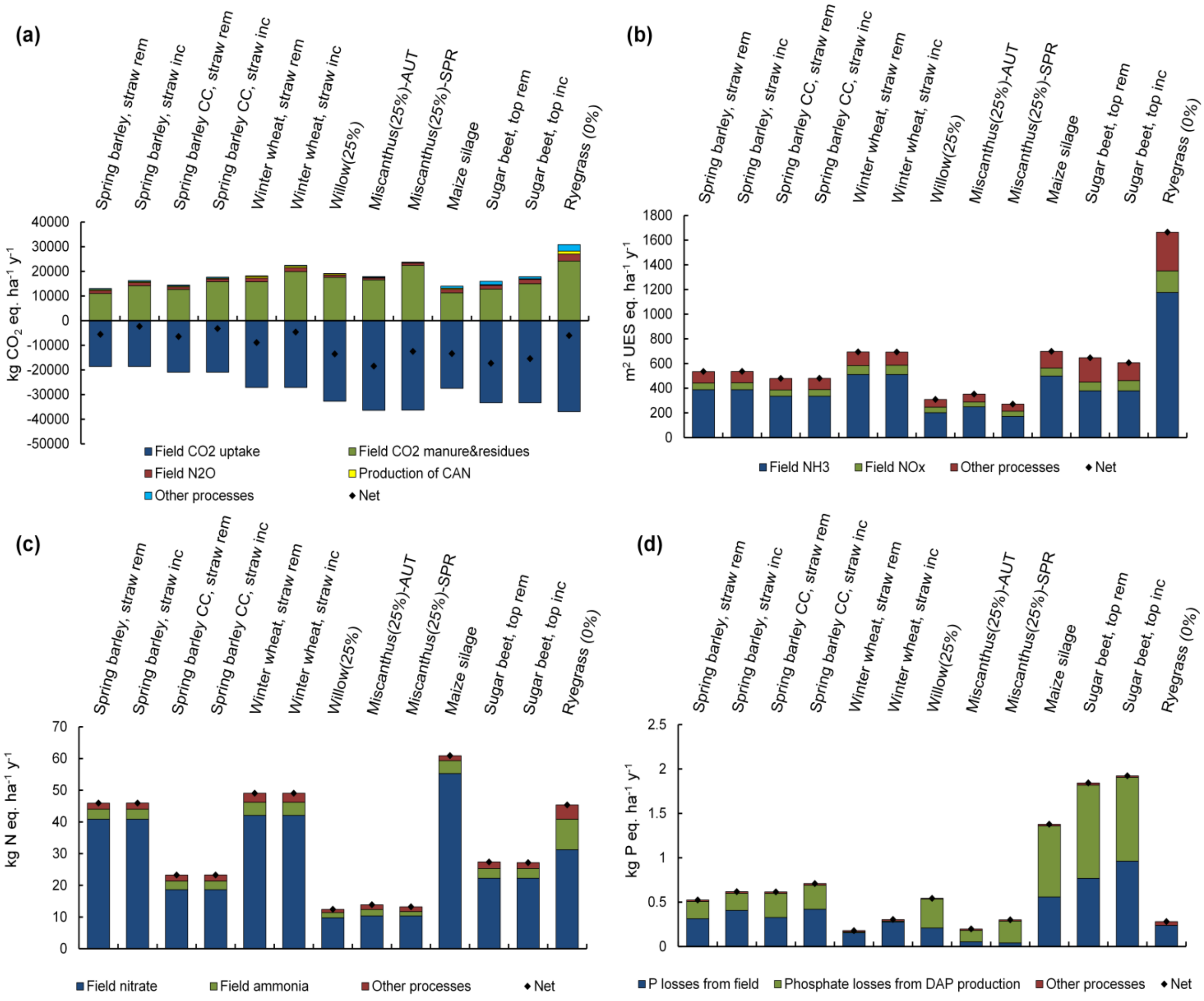

Figure 12. Cradle-to-gate LCA results for inventoried crop systems for the following impact categories: global warming (a), acidification (b), eutrophication-N (c), eutrophication-P (d). Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values between parentheses for perennial crops indicate the turnover rate reduction considered because of the absence of tillage. Results are for a sandy soil with a medium initial soil C and under a wet climate.

As shown in Figure 12b, the acidification impact is particularly determined by the NH_3 emissions from the field, and thus, proportional to the amount of N fertilizers applied. Similarly, eutrophication-N (i.e. with N being the limiting nutrient for growth), is essentially determined by the nitrate leaching occurring during cultivation. Eutrophication-P (i.e. with P being the limiting nutrient for growth), on the other hand, is the results of both P losses from the field, and the phosphate losses from the production of DAP. This involves that the more DAP is used, the greater is the eutrophication-P impact. This appears to be an advantage for wheat, which required little DAP input (Hamelin *et al.* ***I***). Overall, Figure 12 also highlights that willow and *Miscanthus* feature net environmental impacts among the lowest for the 4 impact categories considered.

In the aim of validating the LCA database built (i.e. the “DLUC database”), it was compared with other LCA databases. This, however, could only be done for 4 crops, as no life cycle inventories were available for the other crops in the compared databases. The results from this comparison are presented in Figure 13, for the same impact categories used in Figure 12. For the global warming impact, however, the other LCA databases do not include the emissions of biogenic CO_2. For this reason, the global warming results illustrated from the DLUC database (gray shades in Figure 13a) do not include the biogenic CO_2, contrarily to the results presented in Figure 12a.

Although the results are not directly comparable, as the Ecoinvent life cycle datasets are allocated (between crop and straw), and as all datasets represent different countries (indicated within parenthesis in Figure 13), this comparison nevertheless shows that overall, the LCA results obtained from the DLUC database are within the range of those derived from other LCA databases, at least for the 4 impact categories considered (i.e. the most relevant for agricultural systems).

As shown in Figure 13a, this study derived lower global warming results, as compared to other LCA databases, for most crops. One reason for this is that in this study, the N_2O emissions related to the production of nitric acid (needed for the production of CAN) were (conservatively) corrected based on latest available data from European plants (section 2.5). On the contrary, this study displays above-average results for the acidification and eutrophication-N impact categories. Unlike global warming, these categories are site-specific, although the results from the DLUC database tend to be higher than those of the Danish LCAFOOD database as well. In the case of acidification, this is partly due to the lower emission factors used for NH_3 emission in the LCAFOOD database (ca. 0.07 kg NH_3-N kg^{-1} N applied), whereas the inventory performed for the DLUC database considers specific emission factors for the application of pig slurry (0.12 kg NH_3-N kg^{-1} N applied), cattle slurry (0.16 kg NH_3-N kg^{-1} N applied) and CAN (0.02 kg NH_3-N kg^{-1} N applied) (Hamelin *et al.* ***I***), resulting in an overall greater emission factor per kg of N applied. For the eutrophication-P impact, the results obtained in this study can hardly be compared with those of other databases (except for LCAFOOD), since these results reflect the specific Danish N-based legislation, where P may be applied in excess (section 7.7). The results from the DLUC database differs from those of the LCAFOOD database as the former considered a crop-dependent emission factor for P losses (5% of the surpluses for annual crops), while the latter considered a soil-dependent fixed emission factor (23-31 kg P losses on sandy loam soils; 53-77 kg P losses on sandy soils), where losses can occur even if no crops are cultivated (LCAFOOD, 2000).

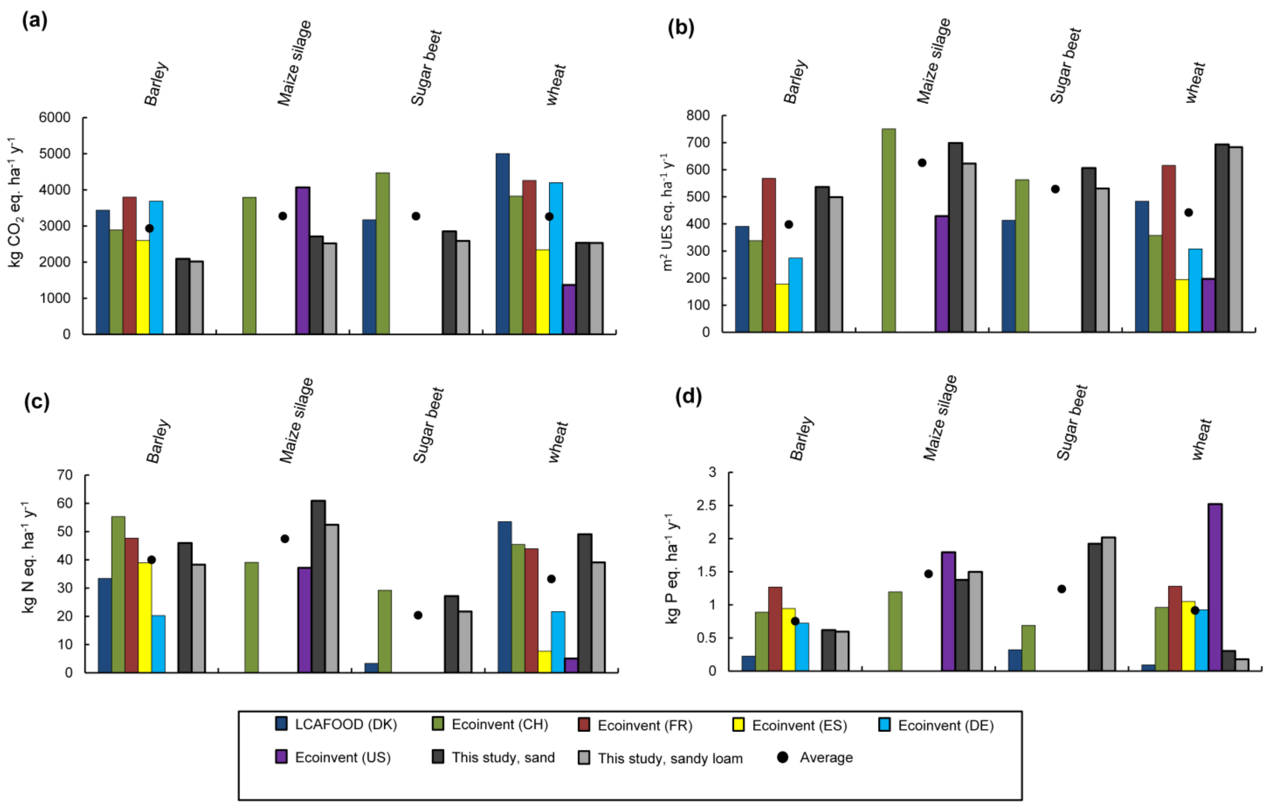

Figure 13. Comparison of the LCA results obtained in this study to those obtained from other available LCA datasets, for the crops for which this was possible. The country represented by each dataset is indicated between parentheses. Results are compared for the following impact categories: global warming (a), acidification (b), eutrophication-N (c), eutrophication-P (d). Results for “this study” consider the incorporation of the secondary yield for barley, wheat and sugar beet, and are for a sandy soil with a medium initial soil C, under a wet climate. Averages include all results displayed in the graph.

7. INCLUDING MANURE INTO LCAS

7.1 Challenges related to manure in LCAs

One challenge with manure systems is that the emissions at any point of the system will be very much dependent upon the manure composition, which itself undergo several changes throughout the different stages of the system governed by factors such as the management (e.g. handling, treatment, etc.), or site-specific parameters (e.g. temperature). Addressing this dependency upon the composition of the manure throughout the whole manure management continuum has however been very poorly addressed in LCAs so far.

Further, manure management is, in Denmark as well as in most European countries, regulated by some key legislation defining the framework conditions for specific stages of the manure management (e.g. fertilization, storage). In order to model the consequences of any studied alternative involving interactions with the manure system, the framework conditions laid down by these legislations should therefore be taken into account in LCAs.

In this section, a methodology is proposed in order to handle these challenges. The principles described here apply not only for LCAs where manure management is the main objet of the study (e.g. Wesnæs *et al.* 2009; Hamelin *et al.* ***IV***), but for any LCA involving the use of manure (e.g. Tonini *et al.* ***II***).

7.2 Functional unit

Manure is often used as a fertilizer, a source of energy, or both. However, manure is also a waste generated by another sector of activity: i.e. livestock production. In this sense, manure production will not be affected, at least not directly, by a change in the demand for fertilizers or for energy. Therefore, it should be treated as a waste. As a result, the functional unit used to consider manure in LCAs should not be related to the outputs provided by manure, but rather be input-based. For example, the functional unit used in Hamelin *et al.* (***IV, V***) was the management of 1 ton of manure, as excreted.

7.3 Manure continuum

The term manure refers to the excreta[53]-based waste produced from animal production systems. It may be solid (can be stacked in a heap), semi-solid (cannot be pumped nor stacked in a heap) or liquid (can be pumped and flow under gravity). Liquid manure with a DM content below 10% is also referred to as slurry (Pain & Menzi 2011). The LCAs performed within this PhD work encompassed slurry only, this representing more than 85% of the manure produced in Denmark (Figure 3).

The prevailing manure/slurry management continuum, in Denmark as well as in most European countries, may be summarized as four main system stages, at which a technology/intervention can be applied: I) feed and feeding systems, II) housing systems including in-house manure management and storage, III) outdoor manure management and storage, and IV) field application of manure. This is illustrated in Figure 14 (reproduced from Hamelin & Wenzel, ***III***).

[53] This includes feces and urine.

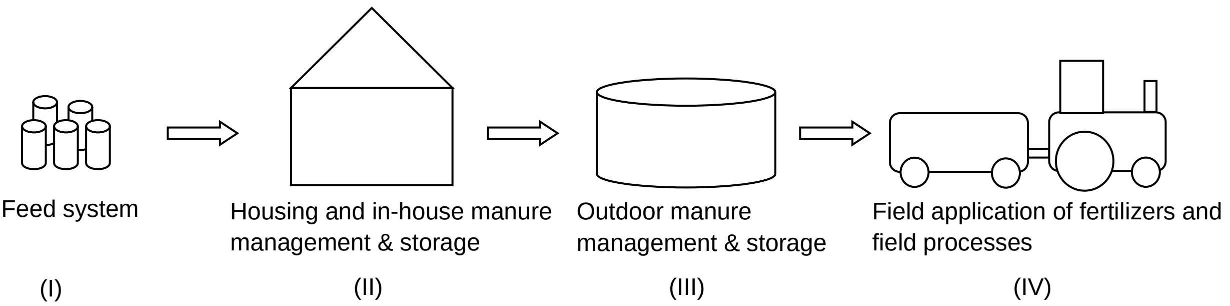

Figure 14. Manure management continuum (reproduced from Hamelin & Wenzel, ***III***)

In the feed system, the different crop (e.g. wheat, barley, soy, rape) and non-crop ingredients (e.g. enzymes, mineral supplements) are produced (stage I) and fed to the animals (stage II). The portion of the consumed feed not respired, evaporated or retained by the animals will end up as excreta (urine or feces). Any change in the feed system (e.g. change of any ingredient) would thus affect the composition of the excreted manure.

Except for grazing or specific production systems (e.g. free range, organic production), urine and feces are generally excreted inside animal houses, and form slurry once they are mixed together (and eventually with water, e.g. spillage and/or washing water) (stage II). Typically, this freshly produced slurry is stored in a slurry pit, beneath the animals (stage II). Depending on the specific management practices, the slurry can remain in-house for varying periods of time: i.e. anywhere from a very short period (e.g. less than a day) to being stored during the whole animal production duration (more than a month). In cases where slurry is not stored in-house for the whole production duration, it is transferred from the animal house towards an outdoor storage facility, where it can be stored (stage III) until its use as a fertilizer (stage IV).

All system stages are characterized by emissions to the environment, and these, in turn, induce changes in the manure composition. As manure composition is the basis for any emissions flows from the manure system, a changed manure composition at one stage of the continuum will in turn trigger changes in the emission flows occurring at other stages of the continuum. For this reason, a procedure allowing to reflect the changes in manure composition at all stages is necessary when dealing with manure systems in LCAs.

7.4 System boundaries issues

Not all stages of the continuum illustrated in Figure 14 have to be included in the LCA, if they are not affected by the alternatives studied. For example, the feed system was not included in Hamelin *et al.* (***IV, V***), as it remained unaffected in all alternatives investigated. Hamelin & Wenzel (***III***) however provide examples where the feed system should be included. Similarly, the animal production system is rarely affected by manure management alternatives, although it could be (Hamelin & Wenzel, ***III***), in which case it would have to be included in the LCA.

7.5 Establishing a reference system

As mentioned in section 2.2, consequential LCAs strive to reflect the environmental consequences of changes. By definition, a change involves two states: one before the change and one after the change. The former represents the reference system, i.e. the point of departure against which the studied alternatives (the change) can be compared.

7.5.1 Step-wise procedure

Defining this reference therefore constitutes a key step in performing LCAs. Depending on the geographical scope of the assessment to be carried out, the reference system may represent a particular farm, a region, a country or even a set of countries (e.g. Nordic countries). As earlier mentioned, the purpose of the reference is to ensure a common ground for the assessment and quantification. As a result, it cannot cover all situations and possibilities, but simply be representative of a "typical" system.

For the specific case of manure management, defining a reference system is not a straight-forward task, due, again, to the “biological” nature of the manure. In this PhD work, a step-wise procedure comprising four main steps was established in order to do so. These steps are briefly described below:

1. *Step 1: Describing the technological status of the reference manure management:*

   This step consists of identifying and describing the technological status of the managerial practices of the reference system that can influence the emissions to environment (referred to as the “managerial reference”, in Hamelin & Wenzel ***III***). Key aspects to describe include:

   - Housing system
     - Which type of floor system is used?
     - How is the manure stored in the housing system?
     - How long is the manure stored in-house and at which temperature?
   - Outdoor storage
     - Describe the storage facility (e.g. concrete tank, earthen lagoon, stack lying directly on the ground, etc.)
     - Is the storage covered? If so, how?
   - Field
     - How is the manure applied to field (e.g. broad spreading, trail hoses, incorporation, etc.)
     - When in the year is the manure spread?
   - Transportation of manure
     - Is it, at any point of the system, necessary to transport manure? If so, what is the transportation distance?

   The above-listed aspects are strictly those whose determination was found necessary when establishing the manure management references described in Hamelin et al. (***IV, V***). It is however acknowledged that additional aspects may be necessary to define. For example, the data or estimation methodologies available for some emission flows (step 4 below) may require the knowledge of other parameters (e.g. ventilation flow). Additional aspects in describing the technological status of the reference may also be determined by the specific object of the study. For example, if the assessment includes the comparison of feeder alternatives, the description of the feeder system is of course an imperative.

2. *Step 2: Describing the site-specific conditions*

   This step involves determining key information related to the geographical scope of the system studied, among others:

   - Average annual temperature;
   - Establishing a “reference crop rotation” (including NPK needs) on which the manure will be applied[54]. An example of such reference crop rotation is given in Hamelin *et al.* (***IV, V***);
   - Types of soil on which the manure will be applied;
   - Description of all relevant legislation governing the manure management practices.

[54] Often, the system boundary will not include the crop system. Nevertheless, it is generally necessary to define a crop rotation in order to assess particular flows (e.g. nitrate leaching, phosphorus losses).

3. *Step 3: Establishing a reference manure composition (in parallel with step 4)*

   Since the emission flows of the manure continuum system are essentially dependent upon the manure composition, it is necessary to set a concrete manure/slurry composition when including manure into LCAs. It is not crucial (either possible) that this manure composition is "correct", rather it should be a fair representation of a manure deriving from the reference situation in question. Key parameters of the reference manure include the dry matter, nitrogen (inorganic and total), phosphorus, potassium, carbon and volatile solids content of the manure. The reference manure composition shall be established for all main stages of the manure continuum:

   - Manure ex-animal: This is the "fresh" manure as excreted by the animals, i.e. prior to any losses to air or to any addition.
   - Manure ex-housing: This is the manure as it leaves the housing units, where it has been stored for a given period of time.
   - Manure ex-outdoor storage: This is the manure as it leaves the outdoor storage.

   The main challenge, when establishing the ex-animal, ex-housing and ex-outdoor storage manure composition is to ensure that consistency is maintained in the mass balances, and that all inputs and outputs (defined in step 4) are systematically and consistently considered. This is illustrated in Figure 15, adapted from Poulsen et al. (2006). Therefore, the task of defining a reference manure composition must be carried out in parallel to step 4 below.

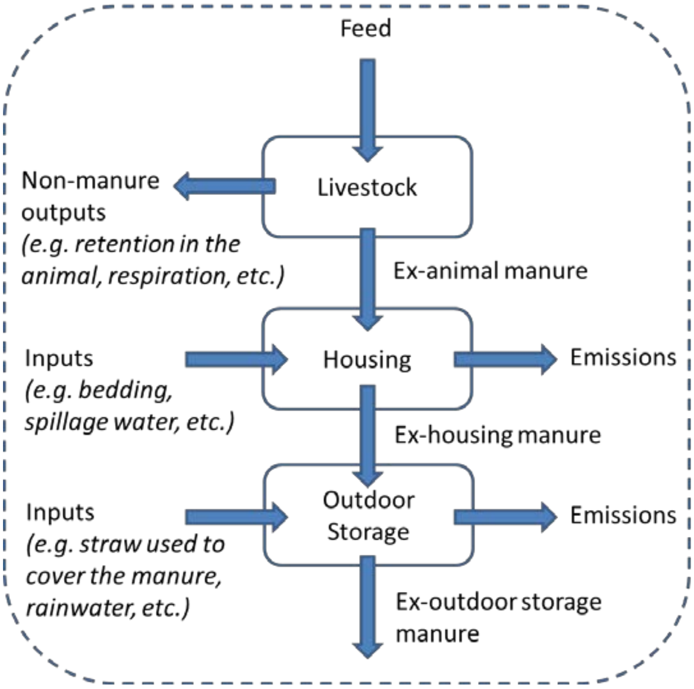

Figure 15. System to consider for establishing a reference manure composition for LCA (adapted from Poulsen *et al.* 2006)

4. *Step 4: Quantifying the key input and output from and to the system (in parallel with step 3)*

   Key input includes:

   - Amount of (rain) water or bedding material added during manure storage (in-house and outdoor) *(in parallel with step 3)*
   - Any significant use of energy (electricity and heat), induced/avoided use of mineral fertilizers, etc.

Key output includes:

- Emissions to air and water compartments throughout the manure continuum *(in parallel with step 3)*
- Emissions from adjoining and background systems (e.g. energy and fertilizer production, use of machinery, etc.).

7.5.2 Danish reference manure management

Based on the above-described step-wise procedure, a Danish manure reference system was established in Hamelin *et al.* (***IV***)[55] for both dairy cow and fattening pig slurry and used in the case studies performed in Hamelin/Tonini *et al.* (***II, IV, V***).

In a nutshell, the reference manure management systems established considered that the produced slurry is stored in-house temporarily (ca. 30 days for dairy cows, 15 days for fattening pigs), and afterwards pumped towards an outdoor concrete storage tank, which is covered. For pig slurry, the cover was considered to consist of a straw floating layer, and for cow slurry, of the natural crust forming on top of the slurry (so no extra cover material is added). When appropriate, the manure is pumped to a slurry tanker and applied on land through trail hoses. Two soil types have been considered, and these are the same as those included in the DLUC database (Hamelin *et al.* ***I***), i.e. a sandy soil (soil JB3 of the Danish soil classification) and a sandy loam soil (soil JB6 of the Danish soil classification, commonly referred to as a “clay soil” within Denmark). Moreover, two reference crop rotations were defined, one on a pig farm and one on a dairy farm. The complete description of the reference manure management established, including all data sources and hypotheses used to define it, is available in the SI of Hamelin *et al.* (***IV***).

For both reference manure management scenarios (i.e. pig and cow slurry), the reference manure composition was established on the basis of the annually updated Danish manure standards (e.g. Poulsen 2011). For this reason, the reference manure composition used in Hamelin *et al.* (***V***) is slightly different than the one presented in Hamelin *et al.* (***IV***), as it was updated on the basis of the latest available values (at the moment of redaction). Further, the estimation methodologies used for estimating some emission flows were also updated. Table 19 presents the reference pig slurry composition considered in Hamelin *et al.* (***V***), while the underlying emission flows are presented in Table 20.

[55] As the study of Hamelin *et al.* (***IV***) is connected to the study performed by Wesnæs *et al.* (2009), the reference system is the same as in Wesnæs *et al.* (2009).

Table 19. Reference slurry composition considered in Hamelin *et al.* (***V***)

Parameter	Slurry ex-animal[a]	Slurry ex-housing[b]	Slurry ex-storage[c]	Source and assumptions
Mass ($t\ pig^{-1}$)	0.47	0.47[d]	0.48	Data needed to ensure correspondence between each manure stage. Values ex-animal and ex-storage based on Poulsen (2011). Value ex-housing based on mass balance[d]. A net water addition of 0.02 m^3 per tonne manure is considered during outdoor storage.
Total N ($kg\ t^{-1}$)	6.00	5.26	5.03	N ex-animal from Poulsen (2011). Losses considered (during housing and during storage): NH_3, N_2O, N_2, NO. Details on N losses are in Table 20. The N from straw addition[e] in-house and as a floating layer during outdoor storage is estimated as 0.009 kg per tonne manure ex-animal and 0.011 kg per tonne manure ex-storage, respectively.
P ($kg\ t^{-1}$)	1.21	1.21	1.19	P ex-animal from Poulsen (2011). No losses considered during housing and storage. The P from straw addition[e] in-house and as a floating layer during outdoor storage is estimated as 0.001 kg per tonne manure ex-animal and 0.002 kg per tonne manure ex-storage, respectively.
K ($kg\ t^{-1}$)	2.83	2.85	2.83	K ex-animal from Poulsen (2011). No losses considered during housing and storage. The K from straw addition[e] in-house and as a floating layer during outdoor storage is estimated as 0.02 kg per tonne manure ex-animal and 0.03 kg per tonne manure ex-storage, respectively.
DM ($kg\ t^{-1}$)	74.8	68.7	66.0	DM ex-storage from Poulsen (2011). Losses during storage: 5 % of the ex-housing values; losses during housing: 10 % of the ex-animal value. Assumptions for losses during storage and housing based on Poulsen (2008).
VS ($kg\ t^{-1}$)	60.7	54.6	52.1	VS are assumed to constitute 79 % of the DM content. Losses considered during storage and housing (absolute values) are the same as for DM (i.e. it is assumed that all DM lost was VS).
C ($kg\ t^{-1}$)	34.5	34.2	31.6	C ex-storage = 47.9 % of DM ex-storage for pigs, based on the ratio C: DM obtained by Knudsen & Birkmose (2005). Losses considered (during housing and during storage): CH_4 and CO_2. Details on C losses are in Table 20. The C from straw addition[e] in-house and as a floating layer during outdoor storage is estimated as 0.75 kg per tonne manure ex-animal and 0.95 kg per tonne manure ex-storage, respectively.
Cu ($g\ t^{-1}$)	31.0	31.0	30.4	Cu ex-storage = 0.0453 % of DM ex-storage, based on the ratio Cu: DM obtained by Knudsen & Birkmose (2005). No losses considered during housing and storage. The Cu from straw addition[e] in-house and as a floating layer during outdoor storage is estimated as 4.92 mg per tonne manure ex-animal and 6.25 mg per tonne manure ex-storage, respectively.
Zn ($g\ t^{-1}$)	90.8	90.7	89.1	Zn ex-storage = 0.135 % of DM ex-storage, based on the ratio Zn: DM obtained by Knudsen & Birkmose (2005). No losses considered during housing and storage. The Zn from straw addition[e] in-house and as a floating layer during outdoor storage is estimated as 75.5 mg per tonne manure ex-animal and 95.9 mg per tonne manure ex-storage, respectively.
NH_4-N ($kg\ t^{-1}$)	4.20	3.94	3.07	Value ex-storage based on Poulsen (2011). Value ex-housing assuming 0.75 kg NH_4-N per kg manure ex-housing Poulsen (2008), and value ex-animal assuming 0.70 kg NH_4-N per kg manure ex-animal (EMEP/EEA 2010).

[a] All values of this column are expressed per tonne slurry ex-animal.
[b] All values of this column are expressed per tonne slurry ex-housing.
[c] All values of this column are expressed per tonne slurry ex-storage.
[d] The non-rounded value ex-housing is 0.47089 $t\ pig^{-1}$, and considers a net water addition in-house of 3.57 kg water per pig, the straw addition described in note e) and DM losses shown in this Table.
[e] The N, P and K addition from straw added in the stable considers, based on Poulsen (2008), an addition of 3 kg of straw per animal per year, 3.3 rotations per year, and the above-mentioned amount of manure ex-animal and ex-housing, yielding a total of 0.0019 t straw per tonne manure ex-housing. For the floating layer, the amount considered is based on (Wesnæs *et al.* 2009), i.e. 2.5 kg per tonne manure ex-housing. The straw DM content is 85 % (Møller *et al.* 2000). The N, P, K, Cu and Zn content of straw per kg of DM is 0.00528 kg, 0.0009 kg, 0.015 kg, 3 mg and 46 mg, respectively, based on Møller *et al.* (2000). The C content is taken as 0.4563 kg C per kg DM, based on an average of 13 values from the Biolex database (FORCE Technology 2013).

Table 20. Life cycle inventory data considered for the reference pig slurry management in Hamelin *et al*. (***V***)

Substances	Life cycle stage: in-house *per tonne ex-animal manure*	Life cycle stage: outdoor storage *per tonne ex-housing manure*	Life cycle stage: field *per tonne ex-storage manure*	Comments: in-house	Comments: outdoor storage	Comments: field
NH_3-N	0.714	0.099	0.604	0.17 kg NH_3-N per kg TAN[a] (Poulsen 2008), with 0.7 kg TAN/kg N (EMEP/EEA 2010).	2.5 % of TAN[a] ex-housing (Poulsen 2008); the N ex-housing being estimated according to Poulsen (2008), i.e.: N ex-animal minus NH_3-N losses in-house (and not accounting for other losses).	12% of N applied (Hansen *et al.* 2008) (this is an average for application by trail hose tanker, excluding illegal dates).
NH_3-N, at application			0.015			0.5% of TAN applied, for application by trail hoses, (Hansen *et al.* 2008)
N_2O-N	0.012	0.030	0.050	0.002 kg N_2O-N per kg N ex-animal (IPCC 2006a) (pit storage below animal)	0.005 kg N_2O-N per kg N ex-animal (IPCC 2006a) (liquid/slurry storage)	1% of N applied, (IPCC 2006c)
NO-N (representing NO_x)	1.96×10^{-4}	1.84×10^{-4}	5.03×10^{-3}	0.0001 kg NO per kg TAN ex-animal (EMEP/EEA 2010)	0.0001 kg NO per kg TAN ex-housing (EMEP/EEA 2010)	0.1 × N_2O-N, based on (Nemecek & Kägi 2007)
NO_3-N	0	0	1.68	Leaching from housing systems assumed negligible.	Leaching from housing systems assumed negligible.	Based on Danish $NLES_4$ model (Kristensen *et al.* 2008): SI of Hamelin *et al.* (***V***).
N_2-N	0.013	0.012		0.003 kg NO per kg TAN ex-animal (EMEP/EEA 2010)	0.003 kg NO per kg TAN ex-housing (EMEP/EEA 2010)	
CO_2-C	0.36	1.20	31.10	1.83 kg CO_2 per kg CH_4: derived from the Buswell equation (Symons & Buswell 1933) as detailed in the SI of Hamelin *et al.* (***V***).	1.83 kg CO_2 per kg CH_4: derived from the Buswell equation (Symons & Buswell 1933) as detailed in the SI of Hamelin *et al.* (***V***).	Based on Danish C-TOOL model, 98.3% of the C applied end up as CO_2-C, over 20 y: SI of Hamelin *et al.* (***V***).
CH_4-C	0.54	1.80	0	IPCC (2006a) algorithm: SI of Hamelin *et al.* (***V***).	IPCC (2006a) algorithm: SI of Hamelin *et al.* (***V***).	Assumed negligible.
P leaching	0	0	0.060			5% of surplus, based on Hamelin *et al.* (***I***).
indirect N_2O-N (volatilization)	7.14×10^{-3}	9.91×10^{-4}	6.09×10^{-3}	1% of N loss as NH_3 and as NO_x, (ex-animal) (IPCC 2006c)	1% of N loss as NH_3 and as NO_x, (ex-housing) (IPCC 2006c)	1% of N loss as NH_3 and as NO_x, (ex-storage) (IPCC 2006c)
indirect N_2O-N (leaching)	0	0	0.013	0.75% of N lost through leaching (in manure ex-animal) (IPCC 2006c)	0.75% of N lost through leaching (in manure ex-animal) (IPCC 2006c)	0.75% of N lost through leaching (in manure ex-animal) (IPCC 2006c)

[a] Ammonium-N (NH_4^+-N) and compounds readily broken down to NH_4^+-N are referred to as total ammoniacal N (TAN).

7.6 Overview of emission factors and estimation methodologies

In Hamelin & Wenzel (***III***), an overview of the emission factors and estimation methodologies available for estimating the emission flows related to manure management (step 4 of the step-wise procedure for establishing a reference manure management system) is provided, in addition to the estimation methodologies already presented in Table 20.

7.7 Modeling fertilizer substitution in LCA – case of Denmark

When farmers apply manure on land, mineral fertilizers (the marginal ones, section 2.5) are substituted. The rationale is that without the manure, mineral fertilizers are what farmers would apply to fulfill the fertilization needs of their crops.

When determining how much mineral fertilizers are actually avoided by 1 kg of N, P and K from the manure, two main parameters must be considered: i) the applicable fertilization legislation; ii) determining if some nutrients are applied in excess (any nutrient applied in excess cannot be considered as "mineral fertilizer avoided").

7.7.1 Applicable fertilization legislation

Based on the Danish legislation (Danish Ministry of Food, Agriculture and Fisheries 2012), there is a maximal amount of N that can be applied to an agricultural field, depending on the crop and soil types, among others. These quotas, per crop and soil type, are updated yearly by the Danish Ministry of Food, Agriculture, and Fisheries (e.g. Danish Ministry of Food, Agriculture and Fisheries 2009b).

However, when the N applied comes from organic fertilizers like manure, not all the applied N will, during the growing period, end up in an inorganic (or mineralized) form that plants can use. To acknowledge this, the law fixes so-called "N utilization efficiency factors" for a variety of manure types, which are 75% for pig slurry and 70% for cow slurry (Danish Ministry of Food, Agriculture and Fisheries 2012). According to this, applying e.g. 100 kg of pig slurry N is, in fertilization accounts, counted as 75 kg. In other words, these efficiency factors allow farmers to apply slightly more N that prescribed by the above-mentioned quotas, when they use organic fertilizers.

Translated to the LCA need of estimating the amount of mineral fertilizer substituted, this also involves that 100 kg of pig slurry N replaces 75 kg of mineral N fertilizers[56]. Similarly, it was considered, based on the Danish fertilization legislation, that 100 kg of cow slurry N replaces 70 kg of mineral N fertilizers. For digestates consisting of digested pig manure and a co-substrate (e.g. Hamelin *et al.* ***V***), the same substitution value as for pig manure has been used.

The Danish legislation also involves, for livestock farms, a so-called "harmony criteria", which links, for a given farm, the maximal amount of manure that can be applied per ha to the number and type of animals kept on the farm (Danish Ministry of the Environment 2012). In this PhD work, where manure rather than "the farm" was the focus, "land" was considered in terms of soil type and reference crop rotation only and not in terms of which farm it belongs to, under the assumption that any manure, once produced, will end up to be applied on land, somewhere in the country. In this perspective, the "harmony criteria" did not need to be considered in the LCA case studies performed within this PhD work[57]. It is however acknowledged that this assumption may not necessarily always hold true, and that there is an environmental relevance in

[56] It is acknowledged that for a particular field, the utilization efficiency factors defined by law may not necessarily correspond to the reality. However, in practice, it is these that determine how much N will end up to be applied to a particular field. In other words, an extra 33% of the N amount prescribed by the quota will, if fertilized with pig slurry, be applied on the field, and this independently of how much slurry N has really been used by the plants. Therefore, it is the utilization efficiency factors that are considered to determine how much mineral N is substituted.

[57] The "harmony criteria" was considered in Hamelin *et al.* (***IV***), but only for determining how much slurry is to be applied on 1 ha of land, in the perspective of determining the amount of P and K applied in excess. The approach used for this was however refined in Hamelin *et al.* (***V***), where the "harmony" criteria did not need to be considered at all, as described in the text.

considering the distances over which manures have to be transported in order to meet the “harmony criteria”. This, however, was beyond the scope of this PhD work.

Moreover, there are threshold limits on the maximal amount of heavy metals that can be applied to land, when fertilization involves specific waste fractions (e.g. food waste) covered by the Danish legislation regulating the application of waste on land (Danish Ministry of Food, Agriculture and Fisheries 2006). This is relevant for the case study performed in Hamelin *et al.* (***V***), where manure is co-digested with some of these wastes. However, not all the relevant heavy metals have been considered in the mass balances, and it was simply assumed that the various digestates produced did respect the thresholds of the above-mentioned legislation, so the digestates could be applied on land. In practice, this assumption may not necessarily be correct, as further discussed in Hamelin *et al.* (***V***).

7.7.2 Nutrients applied in excess

As opposed to N, there is no quota on the amount of P and K that can be applied on the field, although the Danish Ministry of Food, Agriculture, and Fisheries does prescribe guidelines per crop and soil type for these nutrients (e.g. Danish Ministry of Food, Agriculture and Fisheries 2009b).

This involves that P and K may be applied in excess when fertilization is provided with manure, as manure is dosed on the basis of the N needs only (which are determined by the quota, itself determined by the type of crops cultivated, among others). In this PhD work, only the amount of manure P and K not applied in excess were considered to replace mineral fertilizers. The rationale behind this is that without the manure, farmers would most likely apply mineral P and K up to the crop requirements only, in order to maximize their gross margin.

The calculation basis to determine the portion of P and K applied in excess is detailed in the SI of Hamelin *et al*. (***V***).

7.7.3 Fertilizer substitution, in summary

Fertilizer substitution was modeled as summarized in Table 21.

Table 21. Proportions of marginal N, P and K fertilizers substituted by the organic fertilizers considered in this study

	Proportion of the nutrients applied from organic fertilizers avoiding the production (and use) of mineral fertilizers *(as applied in this study)*	**Comments**
N	75% for pig slurry 70% for cow slurry 75% for digestates consisting of pig slurry and other co-substrates (Hamelin *et al.* ***V***) 65% for urine[a] (Hamelin *et al.* ***V***) 40% for compost (based on neither food waste or manure)	Based on the Danish legislation for fertilization (Danish Ministry of Food, Agriculture and Fisheries 2012).
P and K	In theory varies from 0-100%. Only the portion not applied in excess is considered to substitute mineral fertilizers.	The methodology to quantify the amount applied in excess is detailed in Hamelin *et al.* (***V***).

[a] Source-segregated urine.

7.8 Assessing intervention in the reference manure system

The principles to apply for assessing an intervention in the reference manure system (i.e. a given alternative manure management) are essentially the same as for the reference system: i.e. the same substance flows (and perhaps additional) have to be considered in coordination with the manure composition at the various stages of the continuum, based on a mass balance approach. Examples are provided in Hamelin *et al.* (***IV, V***).

8. MANURE-BASED BIOGAS

8.1 Ambition and challenges related to increased manure-biogas in Denmark

Recovery of manure biogas is a well-known mitigation technology for greenhouse gas (GHG) emissions in agriculture (e.g. Olesen 2005; Cuellar & Webber 2008; Holm-Nielsen *et al.* 2009; Weiland 2010). When designed and operated properly, ensuring e.g. against CH_4 fugitive losses or losses from the digested slurry, manure biogas has in fact been found to be one of the most cost-effective ways of reducing GHG emissions (Olesen 2005; Danish Ministry of Food, Agriculture and Fisheries 2008a), due to simultaneous benefits of reduced CH_4 and N_2O emissions from manure storage and field application as well as of replaced fossil fuels from utilizing the biogas. In the perspective of a fully renewable energy system, biogas also offers the possibility to be storable in the gas network, which provides flexibility for buffering the fluctuant energy supply from intermittent sources like wind and sun (discussed in section 10.1).

In this context, a target has been launched in Denmark to achieve 50% use of manure for biogas by 2020 (Danish Ministry of Food, Agriculture and Fisheries 2009a) as compared to the present use of only 5-7% (Birkmose *et al.* 2013).

There are, however, two major obstacles for a widespread implementation of slurry biogas. First, animal manures are often too dilute, containing too little easily degradable C for ensuring economically attractive CH_4 yields. Further, the supply of N from manure 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 (Hansen *et al.* 1998). These obstacles have traditionally been solved by supplementing the manure with substrates providing additional C input. In Denmark, the strategy used so far has been to use C-rich and easily degradable industrial biowastes (e.g. industrial organic residues from fish, fruit, sugar, dairy or oil industries) as co-substrates. However, the availability of these is rather limited compared to slurry volumes. In fact, the 5-7% of Danish manure being digested nowadays already requires almost all available industrial biowaste, and even involves an import (Danish Ministry of Food, Agriculture and Fisheries 2008b; Jørgensen 2009). In the light of the above-mentioned 50% target, this implies that more than 90 % of animal slurry will need another strategy for increasing the economic feasibility of biogas (assuming, based on Figure 3 and Table 10, that the target will primarily be met by slurry). Such strategies are discussed in the present section, in the perspective of their environmental consequences.

8.2 Key modeling framework conditions used for modeling manure-biogas production in Denmark

This PhD work involves two LCA case-studies investigating the consequences of increased manure-biogas in Denmark. An overview of the key modeling framework conditions considered in these two case studies are summarized in this section.

8.2.1 Functional unit

Recovering biogas from manure is, in this PhD work, viewed as a manure management service, for the reasons explained in section 7.2. In this perspective, the functional unit considered for both LCA case studies is the management of 1 tonne of freshly excreted pig manure (manure ex-animal), however:

- In case study 1, the input to all scenarios is 1 tonne of manure ex-animal, and nothing else;
- In case study 2, the input to all scenarios is 1 tonne of manure ex-animal plus a certain amount of external co-substrates[58].

[58] The amount necessary to get a mixture reaching a DM content of 10% after the first digestion step (section 8.2.3).

8.2.2 Reference system

The reference manure management considered, for both case studies, the same technological status and site specific conditions. The former is as described in section 7.5, and the latter considers Denmark as the geographical scope, i.e. the data inventory for crop cultivation, manure management, and the applicable legislation were based on the Danish context. The reference manure composition, however, present slight variations between the two case studies, reflecting, among others, the application of updates in the second case study.

8.2.3 Biogas production

The biogas production considered in the case studies performed within this PhD work is based on a two-steps anaerobic digestion consisting of a completely stirred main digester and a post-digester from which ca. 10% additional CH_4 emissions are captured. It is assumed that the production is operated under mesophilic conditions, and that the biogas produced is constituted of 65% CH_4 and 35% CO_2[59], with a density of 1.158 kg Nm^{-3} biogas and a LHV of 23 MJ Nm^{-3} biogas[60]. Fugitive CH_4 losses of 1% of the produced CH_4 were assumed, based on recent LCA studies (Börjesson & Berglund 2006; Jungbluth *et al.* 2007; Lansche & Mueller 2012).

The CH_4 yield considered for raw pig slurry is 319 Nm^3 CH_4 ton^{-1} VS, based on original data from Danish biogas plants (Hamelin *et al.* ***IV***). For all scenarios involving co-digestion, co-substrates were added to raw pig slurry in order to get a mixture reaching a DM content of 10% after the first digestion step as well as a C:N ratio limited to 20, reflecting state-of-the-art practices of Danish biogas plants. The calculation procedure to determine the mixture input is detailed in the SI of Hamelin *et al.* (***IV***).

Internal electricity consumption corresponding to 5% of the net electricity production was assumed, based on original data from Danish biogas plants. Internal heat consumption was calculated considering that the mixture is heated from 8°C (Denmark's average annual temperature) to 37°C. Complete details regarding the energy balances of the biogas produced in each alternative assessed are available in the SI of Hamelin *et al.* (***IV, V***).

8.2.4 Biogas utilization

In all alternatives assessed, the biogas was considered to be used for CHP, thereby substituting the production of marginal heat and power. Efficiencies of 46% for heat and 40% for electricity were considered for the biogas engine. In both case studies, it was considered that the net heat produced could not be completely recovered, reflecting the losses occurring in periods with low heat demand (e.g. summer months)[61].

8.2.5 Digestate utilization

The digested effluent resulting from the anaerobic digestion process is here referred to as the "digestate". As for the reference manure management, the digestate is considered to be stored in a concrete tank covered with a straw floating layer. When appropriate, it is applied on agricultural fields as an organic fertilizer, thereby displacing the marginal mineral N, P and K fertilizers. The modeling of fertilizer substitution is further detailed in the SI of Hamelin *et al.* (***V***). Changes in soil C occurring as a result of applying the digestates instead of raw manure were estimated with the dynamic soil C model C-TOOL, the same model as used for establishing the DLUC database (section 6).

[59] 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.
[60] The calculation details for the density and LHV of biogas can be found in Hamelin *et al.* (***IV, V***).
[61] 60% of the net heat produced was assumed recovered in Hamelin *et al.* (***IV***), while a recovery of 90% was considered in Hamelin *et al.* (***V***).

8.3 Case study 1: Manure-biogas from separated slurry (ex-housing)

Separating ex-housing slurry into a liquid and a solid fraction and using the resulting concentrated solid fraction as a co-substrate to raw slurry in anaerobic digesters represents a promising alternative in order to avoid the reliance on constrained C co-substrates for slurry-based biogas. In other words, this concept allows to produce biogas from a mixture of solid fraction and raw slurry and thus to use manure as the only feedstock for anaerobic digestion, and this, based on simple separation technologies already available to farmers nowadays. In Hamelin *et al.* (***IV***), this concept was assessed through 3 scenarios, each considering different slurry separation technologies to obtain the solid fraction input for biogas production. These 3 scenarios were applied to pig slurry and one scenario was applied to both pig and cow slurry. These scenarios, which are thoroughly described in Hamelin *et al.* (***IV***), consist of:

- Alternative P1: Separating the slurry with a conventional centrifugal separation technology in combination with the addition of a cationic polymer, namely polyacrylamide (PAM). This results in a rather high separation efficiency[62] (87% of the DM and 90% of the P ending up in the solid fraction). The liquid fraction obtained from the separation process is stored and used as a fertilizer, while the degassed 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 P management, given the richness of the P content in the degassed slurry, a consequence of the high separation efficiency of the first separation. The resulting degassed liquid and solid 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 (Jørgensen 2004), an increased plant yield was also modeled, as detailed in Hamelin *et al.* (***IV***). This scenario was also applied to dairy cow slurry (SI of Hamelin *et al.* ***IV***).

- Alternative P2: Separating the slurry with a mechanical screw press technology, where the liquid fraction is stored and used as a fertilizer (as in P1). The degassed slurry is not separated as its P content is not high enough to justify a second separation (separation efficiency of 61% for the DM and 66% for P). It is consequently simply stored and used directly as a fertilizer, where an increased crop yield was considered as described for P1.

- Alternative P3: Separating the slurry with a mechanical screw press technology (as in P2), where the produced solid fraction is first used to produce “fibre pellets”. This process consists of drying the solid fraction in a tumble dryer and subsequently pressing it to form pellets with a DM content of 89%, so transportation costs are reduced. It is these pellets that are then 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 K and P fertilizers. The liquid fraction and degassed slurry are dealt with as in P2.

The system boundary considered for all assessed alternatives, including the reference system, is illustrated in Figure 1 of Hamelin *et al.* (***IV***), where relevant mass and energy flows are presented in relation to the functional unit[63]. Moreover, all modeling details are described in Hamelin *et al.* (***IV***).

The LCA results of this case study are illustrated in Hamelin *et al.* (***IV***), with breakdown per process and substance, while the net results for the environmental impact categories addressed are summarized in Table 22. 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 of Hamelin *et al.* ***IV***) from the induced impacts (positive values).

[62] Separation efficiency refers to the proportion of a component (e.g. DM, C, N, etc.) ending up in the solid fraction, expressed as a percentage of the total quantity of this component in the slurry.
[63] The system boundary considered for systems involving dairy cow manure is illustrated in Figure S1 of the SI of Hamelin *et al.* (***IV***).

There is a benefit when the net impact of a given alternative is lower than the net impact of the reference scenario.

Table 22. Net LCA results for the manure-biogas scenarios assessed in Hamelin *et al.* (***IV***)[a]

Impact category	Unit[b]	Pig slurry				Cow slurry	
		Ref-pig[d]	P1	P2	P3	Ref-cow[d]	C1[d]
Global warming (over a 100 y horizon)	kg CO_2 eq. t^{-1} slurry ex-animal	257	**154**	221	242	305	**197**
Acidification	m^2 UES[c] kg CO_2 eq. t^{-1} slurry ex-animal	44.8	41.3	**38.3**	39.4	37.4	**34.9**
Photochemical ozone formation	person ppm h t^{-1} slurry ex-animal	0.165	**0.147**	0.153	0.170	0.144	**0.138**
Eutrophication-N	kg N eq. t^{-1} slurry ex-animal	0.608	0.529	0.532	**0.516**	0.682	**0.640**
Eutrophication-P	kg P eq. t^{-1} slurry ex-animal	-0.00167	**-0.0219**	-0.00951	-0.00551	-0.00631	**-0.0241**
Respiratory Inorganics	kg $PM_{2.5}$ eq. t^{-1} slurry ex-animal	0.238	0.234	**0.210**	0.230	**0.200**	0.202

[a] All values are rounded to 3 significant digits. Bold numbers highlight, for each impact category and slurry type, the alternative allowing the lowest net environmental impacts.
[b] The units are as described in the EDIP methodology: Hauschild & Potting (2005).
[c] Unprotected ecosystem equivalent (described in the EDIP LCIA methodology: Hauschild & Potting, 2005)
[d] Ref-pig: Reference pig slurry management scenario; Ref-cow: Reference cow slurry management slurry; C1: This scenario is as for P1, but involves cow slurry (and thereby different life cycle inventory data, including separation efficiencies)

The main findings of the LCA can be summarized as:

- For all impact categories assessed, all biogas alternatives allowed for a net impact lower or practically equal to what is obtained with the reference manure management system;

- The global warming 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. A greater separation efficiency involves that more easily degradable VS (the degradation of which produces CH_4) are transferred to the solid fraction, which in turn implies a greater CHP production from slurry and thereby a greater displacement of marginal energy;

    - The greatest separation efficiency was obtained for alternative P1, the only alternative allowing for a net global warming impact clearly lower than the one from the reference scenario. Yet, this alternative is dependent upon the use of cationic PAM, which is not free of concerns, as discussed in Hamelin *et al.* (***IV***) and Hamelin *et al.* (2010).

- In-house slurry storage, although it was not affected by the biogas alternatives studied, represented an important contributor to four of the six impacts assessed: global warming, acidification, photochemical ozone formation and respiratory inorganics. Two main substances were responsible for this: CH_4 (global warming and photochemical ozone formation) and NH_3 (acidification and respiratory inorganics). Technologies allowing to reduce both CH_4 and NH_3 emissions from in-house slurry storage were thus highlighted as representing a clear opportunity for improving the environmental performance of slurry management.

- Although obvious, the LCA showed that there are little environmental advantages in using an energy intensive process to dry the separated solid fraction prior to using it for biogas. It is nevertheless acknowledged that there are other advantages related to this pelletization process, for instance reducing the transportation costs of slurry-based fertilizers.

- Results showed eutrophication-P potentials very close to 0 for all alternatives (Table 22), and the breakdown per process highlighted that the P losses from slurry (and slurry-based products) application were counter-balanced by the avoided phosphate emissions related to the production of marginal mineral P fertilizer. The biogas alternatives presented additional benefits compared to the reference slurry due to the avoided marginal electricity and heat production, as these imply releases of phosphate to water.

- Soil C results showed that between 13 and 50% less C ended up in the soil pool with the different biogas alternatives, as opposed to the reference slurry management.

- The processes related to the management of the separated liquid from the first separation (i.e. outdoor storage and field application) accounted for 16-47% of the induced GHG emissions, depending on the scenario. This highlights the importance of including this portion of the slurry by-passing the biogas plant in the LCA; its production is in fact a consequence of applying a biogas production concept based on separated slurry.

Sensitivity analyses were performed and tested with alternative P1 in order to highlight the importance of some of the most sensitive assumptions, including, among others, the soil type on which the slurry-based fractions are applied (sandy loam instead of sandy soil), and the type of marginal electricity (coal instead of mix electricity marginal) and heat (natural gas instead of coal) considered. None of the sensitivity analyses performed resulted in a change of the tendencies presented in Table 22, as further discussed in Hamelin *et al.* (***IV***).

Overall, the results of this case study highlighted the limits of this biogas production concept in the perspective of a wide-spread strategy for increased manure-biogas. In fact, acknowledging that global warming is a key concern, only one alternative allowed for clear GHG reductions compared to the reference slurry management. Yet, this alternative involves the use of a cationic polymer, which likely persists and accumulates in the environment, and which does represent a potential toxicity risk, although this could not be quantified in the LCA. On this basis, further research on efficient separation technologies not involving cationic PAM appears necessary.

8.4 Case study 2: External C addition for increased manure-biogas

Case study 1 investigated the possibility of increasing manure-biogas without relying on the availability of external C co-substrates, and involving technologies that are already available to Danish farmers nowadays.

In case study 2, additional options were investigated, with a focus on external C co-substrates. Five external co-substrates not already fully used for biogas were considered: energy crops, straw, household biowaste, commercial biowaste and garden waste. Further, the use of the solid manure fraction deriving from source-segregation of animal urine and feces was also investigated. The latter option differs from case study 1, as it involves a separation system directly under the animals, where the contact between feces and urine is prevented at first place. The substrates chosen were those considered to have the greatest potential to supply an increased manure-biogas production. Substrates already fully used for the manure being digested nowadays (i.e. industrial biowastes) were thus not considered.

These co-substrates, however, already have their uses/disposal routes, and using them for biogas diverts them from their initial use or disposal route, here referred to as the “lost alternative”. These lost alternatives imply a variety of environmental and economic consequences, among others the production of a substitute to supply the service (e.g. energy, fertilizer) no longer provided by the co-substrates. In Hamelin *et al.* (***V***), the environmental consequences of this were assessed in the perspective of establishing a sustainable strategy for achieving a colossal increase in manure-biogas in Denmark.

Seven baseline co-digestion scenarios were analyzed, where the co-substrates were digested together with 1 tonne of raw pig slurry (ex-animal) (functional unit)[64]. The amount of co-substrate added to this was determined in order to get as close as possible to the 10% DM threshold fixed[65] after the first digestion step (section 8.2.3). In all scenarios, raw slurry was transferred to a biogas plant right after the housing stage, biogas was used for CHP production and the digestate was stored and used as a fertilizer, thereby replacing marginal mineral N, P and K, as described in section 7.7. The assessed scenarios are briefly described here, but a more thorough description can be found in Hamelin *et al.* (***V***):

- Scenario 1, Energy crops: Maize silage has been chosen as the energy crop to represent this scenario given its high yield and its high C turnover efficiency (Hamelin *et al.* ***I***; section 6). It is considered to be produced in Denmark specifically for anaerobic digestion, and as such is displacing another crop (DLUC), which is here considered to be maize for animal feed. This reduced supply of Danish feed maize will, in turn, induce ILUC, which was modeled as described in section 4.3, although the environmental consequences of intensification, displacement and foregone C sequestration were not accounted for. Modeling details related to ILUC are further described in the SI of Hamelin *et al.* (***V***). Once harvested, the maize is temporarily stored prior to its use for co-digestion.

- Scenario 2, Straw: Winter wheat straw is the most abundant straw in Denmark (Statistics Denmark 2012d), and was therefore the straw type considered for this study. As for the maize scenario, straw is harvested and stored temporarily. Prior to co-digestion, straw undergoes an extrusion pre-treatment (described in Hjorth *et al.* 2011), allowing to break straw's lignocellulosic structure and render a maximum of its C content bioavailable. If not used for co-digestion, it was considered that straw would have otherwise been combusted in a small-to-medium scale biomass CHP plant, where it would have produced heat and electricity. This involves that an equivalent amount of energy from the marginal heat and electricity suppliers is induced in the system in order to replace the energy no longer provided as straw is diverted for biogas.

- Scenario 3, Household food waste: This scenario involves the use of the food waste generated every year from the Danish households, also termed biowaste. Once collected, biowaste is separated from the overall household solid waste through a press separation technology. Prior to co-digestion, the biowaste undergoes, as required by EU legislation (European Union 2011), a hygienization process, where it is heated from 8°C to 75°C. If not used for biogas, it was considered that household biowaste would have otherwise ended in a municipal solid waste incineration CHP plant, where it would have produced heat and electricity. As for straw, the energy that is no longer produced because household biowaste is now used for biogas was modeled as marginal heat and power induced.

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[64] It is however the manure ex-housing that is used as an input for the anaerobic digestion process. Based on the reference manure composition established for fattening pig slurry (Table 19; see footnote d), there is 1.002 t manure ex-housing per t manure ex-animal.

[65] This 10% limit has been empirically determined based on decades of experience in Danish biogas plants, and is used in practice to dose the input mixture to digesters in cases where the input mixture is dominated by manure (personal communication with Anders Peter Jensen, Xergi, October 2011). It mostly reflects the performance of the pumps and mixers in the digester. Therefore, it should not be seen as a universal threshold, neither as a threshold limit for the biological performance. A different threshold would likely be used for mixtures based on a different substrate than manure.

- Scenario 4, Commercial biowaste: This scenario considers commercial biowaste (or food waste) from wholesale and retail stores. The same life cycle processes as for the household food waste scenario were applied for this scenario.

- Scenario 5, Garden waste: Garden waste (which is also a biowaste) is generated during maintenance of public areas and private gardens. In this study, garden waste is assumed to be constituted of 75.6% easily degradable material (e.g. leaves and grass) and 19.5% branches, the rest consisting of woody parts, stones and foreign items, based on the findings of Boldrin & Christensen (2010). Before to be fed to the digester, garden waste is shredded. The lost alternative considered for garden waste is open windrow composting, which would have produced two outputs: screened wooden materials burned in small-to-medium scale CHP plants and mature compost applied on land as a fertilizer. The energy and fertilizing services no longer provided as the garden waste is diverted to biogas has for consequence that equivalent amounts of marginal energy and fertilizers are induced.

- Scenario 6, source-segregated manure: In this scenario, 1 tonne of raw pig slurry is co-digested with the concentrated solid fraction obtained from source-segregation of urine and feces in the animal house (of a second farm). This involves that urine and feces are not mixed together in this second farm. The separated solid and liquid fractions are stored temporarily in-house (less than 1 week), and the liquid is then stored and applied on land, thereby replacing marginal mineral fertilizers. If not used for co-digestion, it is considered that the manure from the second farm would never have been separated, but merely managed according to the reference manure management. This is thus the lost alternative considered.

- Scenario 7, mono-digestion: A mono-digestion scenario was included for comparison purposes only. In this scenario, 1 t of raw pig slurry (ex-housing) is digested on its own, which avoids the reference slurry management to occur.

All modeling parameters related to these scenarios (e.g. CH_4 yields, emissions, mass balances, LHV of co-substrates, etc.) are thoroughly detailed in the SI of Hamelin *et al.* (***V***). Further, the system boundary conditions, where relevant mass and energy flows are presented in relation to the functional unit, are illustrated in the SI of Hamelin *et al.* (***V***) (Figures S1-S7) for all other scenarios.

Sensitivity analyses were performed on the above-described scenarios in order to highlight the importance of some of the most sensitive assumptions, without repeating what has already been assessed in case study 1. A total of 11 additional scenarios were thus performed, consisting of:

- Different lost alternatives for selected scenarios, namely straw plowing (instead of combustion) for the straw scenario, landfilling (instead of combustion or composting)[66] for the three biowastes scenarios, and mono-digestion (instead of conventional manure management) for the source-segregation scenario (i.e. assuming that if not separated, the manure from the second farm would have been mono-digested instead of being handled according to the reference manure management) *(6 scenarios)*;

- Different variants for the energy crop scenario, namely that (i) spring barley and (ii) sugar beet were displaced instead of feed maize. One additional variant (iii) for maize was to consider a natural on-field drying prior to harvest, so its DM content could rise from 31% (baseline case) to 40%, thereby allowing to reach a mixture manure-maize with 10% DM (as opposed to 6.6% in the baseline scenario) *(3 scenarios)*;

[66] For garden waste, both landfilling and combustion were considered (instead of composting).

- Different pre-treatments for the straw scenario namely an alkali pre-treatment as well as a pre-treatment combining straw explosion and enzymatic hydrolysis (instead of the baseline extrusion pre-treatment) *(2 scenarios)*.

Each scenario required different amount of co-substrates per tonne of fresh manure excreted (functional unit), and consequently, different quantities of biogas were produced for each of them. This is shown in Table 23.

Table 23. Co-substrates required, biogas produced and input mixture characteristics of each alternative assessed in Hamelin *et al.* (***V***).

	Energy crop	Straw	Household food waste	Commercial food waste	Garden waste	Souce-segregated manure	Mono-digestion
Co-substrates required per FU (kg wet weight)[a]	1303	183	785	1701	297	1441	-
Biogas produced per FU (Nm^3)	250	87	131	193	75	194	27
C/N ratio of input mixture	19.7	17.1	11.9	13.2	13.1	10.1	6.5
DM of mixture after 1st digestion step (%)	6.6	10.0	10.0	10.0	10.0	10.0	3.7
Share of manure in input mixture (%)	43	85	56	37	77	100[b]	100
Nm^3 CH_4 produced per t mixture	70	48	48	46	38	52	18

[a] This consists of the silage maize as leaving the storage, the straw as leaving the extrusion pre-treatment, the household and commercial biowastes as leaving the hygienization process, the garden waste as leaving the shredding process and the source-segregated manure as leaving the in-house storage.
[b] Consisting of 41% manure ex-housing and 59% source-segregated solid fraction.

The LCA results for the baseline and sensitivity analyses are illustrated in Figures 1 and 2 of Hamelin *et al.* (***V***), with breakdown per process. The net results for all baseline and sensitivity analysis scenarios are presented in Table 24 and Table 25, respectively. The net impact for a given alternative is obtained by subtracting the avoided impacts (negative values; these are environmental benefits) from the induced impacts (positive values). Results are expressed per functional unit (FU) for all impact categories, although for global warming, results were also expressed per Nm^3 biogas produced, and per ton DM input.

Table 24. Net LCA results for the baseline scenarios, for the case study performed in Hamelin *et al.* (***V***). Values are rounded to 2 significant digits. Bold numbers highlight the scenarios where the environmental net impacts are the lowest

Scenarios	Global warming (kg CO_2 eq.) Per FU[a]	Per Nm^3 biogas	Per ton DM input	Acidification (m^2 UES per FU[a])	Eutrophication-N (kg N eq. per FU[a])	Eutrophication-P (kg P eq. per FU[a])
Baseline						
Energy crops	1000	4.1	2.2	36	2.6	8.1×10^{-2}
Straw	-140	-1.7	-0.64	10	0.33	3.1×10^{-3}
Household food waste	-101	-0.78	-0.32	36	1.6	1.7×10^{-2}
Commercial biowaste	-32	-0.17	-0.067	50	3.0	2.9×10^{-2}
Garden waste	-310	-4.2	-1.3	13	0.23	$\mathbf{-1.4 \times 10^{-4}}$
Source-segregated manure	**-1300**	**-6.5**	**-2.7**	**-110**	**-0.71**	8.2×10^{-3}
Mono-digestion	-130	-4.8	-1.9	18	0.16	6.8×10^{-5}

[a] FU: Functional unit, which is here 1 tonne of manure ex-animal.

Table 25. Net sensitivity analysis results for the global warming impact, for the case study performed in Hamelin *et al.* (***V***). Results in kg CO_2 eq. per functional unit.

	Lost alternatives					Variants for the energy crop and straw scenarios				
	Straw, Plowing	Household food waste, landfill	Commercial biowaste, landfill	Garden waste, incineration	Garden waste, landfill	Energy crop, displacing barley	Energy crop, displacing sugar beet	Energy crop, reaching 10% DM (on-field drying)	Straw, alkali pre-treatment	Straw, straw explosion + enzymatic hydrolysis pre-treatment
Sensitivity analysis results	-460	-180	6.2	140	160	-140	1800	3200	-110	-80
Baseline results	-140	-101	-32	-310	-310	1000	1000	1000	-140	-140

The main findings of this case study can be summarized as follows:

- Digesting manure instead of managing it on the basis of the reference manure management yields significant environmental benefits, for all impact categories. This translated to an important additional benefit for the scenarios allowing to use more manure for co-digestion.

- “Energy crop” was the co-substrate scenario allowing to produce the greatest amount of biogas per functional unit. However, it also had the highest environmental cost for most impact categories assessed, in particular global warming. This result is mostly due to the ILUC it involved.

- Source-segregated solid manure was highlighted as the co-substrate yielding the greatest environmental benefits overall. This mostly reflects that it allowed to use a lot more manure for biogas than the other scenarios, so more “reference manure management” was avoided.

- Except for the source-segregated manure scenario, global warming was the only impact for which significant reductions could be achieved through the biogas scenarios. For the other impact categories assessed, the main hot spot was the field application of the digestate. The main substances responsible for this were NH_3 (acidification), nitrate losses (eutrophication-N) and P losses (eutrophication-P), respectively. Yet, the emissions of NH_3 from field application of the digestate (and liquid fraction) could be significantly reduced through the application of additional mitigation technologies (e.g. acidifying the digestate; Wesnæs *et al.* 2009[67]), which could render the net acidification impact close to zero for all scenarios (except, of course, source-segregation, which is already below zero). Similarly, P losses could be minimized through precision dosing, i.e. by applying the digestates with high P content (Table S64-S65 of the SI of Hamelin *et al.* ***V***) in areas with P deficits, while the N losses could be reduced through e.g. the use of additional catch crops in the crop rotation.

- Straw and biowastes (i.e. garden waste as well as household and commercial food waste) all allowed a net GWP reduction, which highlights that GHG-wise, these co-substrates are better used in biogas than in their previous use (i.e. composting for garden waste and combustion for straw and the other biowastes). It was also highlighted that co-digestion of straw and biowastes with manure has two additional advantages over incineration: i) it allows to recycle these co-substrate’s nutrients

[67] In Wesnæs et al. (2009), the principles of the “manure acidification technology” are described and a LCA is performed for the case of raw slurry acidification (but not digested manure, although the same principles would apply).

and slowly degradable C, which are essentially lost in the incineration case, and ii) it produces a storable gas, a key flexibility asset for a renewable energy system.

- The sensitivity analyses (performed for the global warming impact only) highlighted that the lost alternative for straw and biowastes had significant importance on the absolute results, particularly for straw and garden waste. For example, it showed that the benefit of the straw scenario could be ca. 3 times higher if straw plowing was avoided instead of combustion. Similarly, for the source-segregated manure scenario, if the lost alternative for the farm providing the separated manure would have been to use this manure for mono-digestion (which would be realistic in a future where all manure are digested) instead of the reference manure management, much less savings, in terms of GWP, would be obtained from this scenario (although savings would still be obtained).

- The sensitivity analyses performed on the crop displaced by more maize silage in Denmark (DLUC) showed that displacing barley instead of feed maize would result into net savings, while displacing sugar beet instead of feed maize would result into a net impact greater than the one observed in the baseline scenario. In both cases, this reflects the differences in yield for these crops. For sugar beet, whose yield is ca. 3% greater than maize, the dominant effect is the "DLUC" one, i.e. the overall GHG emissions of the cultivation system are smaller for sugar beet than maize, as shown in Figure 12a. For barley, whose yield is ca. 180% lower than maize, the main effect is however the "ILUC" effect, i.e. it reflects that as less crops (and thus carbohydrates) are then displaced, less land conversion is needed to replace the carbohydrates no longer supplied from Denmark. However, in the perspective of a high bioenergy future like the one modeled in Hamelin *et al.* (***V***), it is likely that there will quickly be no more low-yielding crop like barley to offset, in which case this "low-yield displaced crop benefit" could not be applied.

- As it could be expected, the sensitivity analysis considering a natural drying of the maize to 40% DM had the effect to increase the share of maize in the input mixture, thereby increasing the energy produced, but also leading to a greater ILUC. This thus resulted in a greater net global warming impact than in the baseline energy crop case.

The implications of these findings are discussed in section 10.

9. FROM CROPS TO HEAT AND ELECTRICITY: COMPARISON OF THE CONVERSION TECHNOLOGIES

9.1 Context

Previous chapters highlighted that the use of energy crops for bioenergy production has an incidence on land use changes that may, in turn, imply C releases greatly exceeding C savings. However, section 6 as well as Hamelin *et al.* (***I***) also pointed out that not all energy crops are equal, and that some energy crops, in particular perennial crops, do have the potential to minimize the drawbacks associated to land use changes.

Beyond energy crops, the type of conversion technology used to produce bioenergy can also influence the total C (and N) balance. In the perspective of identifying the most sustainable *crop* × *technology* combinations, the LCA case study carried out in Tonini *et al.* (***II***) assessed the environmental impacts associated with the production of bioenergy (as heat and electricity) from 1 hectare of Danish arable land cultivated with 3 different perennial crops, i.e. ryegrass, willow and *Miscanthus* (spring harvested). For each, four different biomass-to-energy (BtE) conversion pathways were considered: i) anaerobic co-digestion with manure, ii) gasification, iii) combustion in small-to-medium scale biomass combined heat and power (CHP) plants and iv) co-firing in large scale coal-fired CHP plants. A total of 12 scenarios have therefore been assessed.

9.2 Key modeling framework conditions

9.2.1 Scope and functional unit

As mentioned above, the FU upon which all input and output flows were expressed was 1 ha of agricultural land used to grow the selected energy crops. As for previous case studies, the geographical scope considered was Denmark (so the data inventory applied for e.g. crop cultivation, BtE plants, legislation, etc. were specific to Denmark). However, the temporal scope considered was 20 years, i.e. all assessed systems were operated for a 20 years duration.

9.2.2 Overview of the system boundary

For all scenarios, thus, 1 (extra) ha of Danish land was cultivated with whether *Miscanthus*, willow or ryegrass, and bioenergy was produced from the biomass that could be harvested on that 1 ha. The inventory data used for crop cultivation were based on Hamelin *et al.* (***I***), for a sandy loam soil and a wet climate. Once harvested, crops were stored (dry storage), pre-treated[68], and fed to the BtE considered. The heat and electricity then produced were displacing the marginal heat and electricity fuels (section 2.4). In the anaerobic digestion case, the digestate was used as a fertilizer on agricultural land, thereby preventing the production and use of mineral N, P and K fertilizers.

As earlier mentioned (section 2.6), the LCA system established in this case study considered that the land used for cultivating the energy crops would have otherwise been used for cultivating spring barley. The DLUC consequence of this translated into the environmental impacts of cultivating the selected energy crops instead of spring barley. The ILUC resulting from this reduced supply of Danish barley was modeled as described in section 4.3 (land expansion only).

The anaerobic digestion scenarios involved co-digestion with fattening pig slurry. As in section 8, it was considered that using this slurry for biogas prevented the “reference manure management” to happen. The same reference manure management (including manure composition) as established in Hamelin *et al.* (***IV***) was considered. Further, as applied in Hamelin *et al.* (***IV, V***) and as explained in section 8.2.3, the proportion

[68] Only *Miscanthus* and willow combustion did not need a pre-treatment.

of crop and manure in the mixture was calculated in order to ensure a biomass mixture input having a DM content of 10% after the first digestion step.

The three thermal bioenergy scenarios (i.e. gasification, combustion and co-firing) implied negligible residual unconverted carbon that is found in bottom and fly ashes. The bottom ashes were assumed to be used for road construction, substituting for natural aggregates, while the fly ashes were assumed to be utilized for backfilling of old salt mines with negligible environmental impacts.

The system boundaries considered for all scenarios are illustrated in Tonini *et al.* (***II***) (manuscript and SI), where the relevant mass and energy flows are presented in relation to the functional unit. Similarly, details on the modeling parameters (e.g. CH_4 yields, emissions, DM losses, LHV, efficiencies, etc.) are thoroughly detailed in Tonini *et al.* (***II***).

9.3 Pre-treatments

Pre-treatments included on-field drying (ryegrass, for all BtE conversion technologies; willow, for gasification and co-firing), size comminution (10-50 mm; for all crops and all BtE conversion technologies except direct combustion) as well as steam pre-treatment for breaking the lignocellulosic structures of *Miscanthus* and willow undergoing anaerobic digestion. All these pre-treatments are further detailed in the SI of Tonini *et al.* (***II***).

9.4 Biomass-to-energy conversion technologies

The BtE conversion technologies involved in this case study consisted of one biological conversion pathway (anaerobic digestion) and three thermal conversion pathways (gasification, combustion and co-firing). Anaerobic digestion was modeled as described in section 8.2.3. Gasification, a sub-stoichiometric combustion process where biomass is converted to a combustible gas by partial oxidation, was modeled as a fluidized bed reactor. The syngas was assumed to be used in a gas engine yielding the same efficiency as when burning the biogas (Table 26). The cold gas efficiency (CGE)[69] and carbon conversion efficiency (CCE)[70] considered were 70% and 95%, respectively. Combustion was modelled as direct biomass combustion in small-to-medium scale biomass CHP plants, while co-firing was modeled on the basis of a large scale coal-fired CHP plant.

A comprehensive description of all modeling parameters and life cycle inventory data considered is presented in Tonini *et al.* (***II***) (manuscript and SI). A summary of the parameters defining the energy balance of the bioenergy scenarios assessed is presented in Table 26.

[69] The CGE defines the fraction of the feedstock chemical energy (as LHV, dry basis) remaining in the syngas (and not lost as, e.g., heat or in the residue). It is expressed as the ratio between the amount of energy in the syngas (after gas cleaning) and the amount of energy in the biomass (as LHV, dry basis)
[70] The CCE defines the proportion of the feedstock C that is transferred to the syngas (as CH_4, CO and CO_2)

Table 26. Overview of the energy balance and main parameters considered for the bioenergy scenarios assessed in Tonini *et al.* (***II***). Eventual inconsistencies are due to rounding (values rounded to two significant digits)

Parameter[a]		Ryegrass[b]				Willow[b]				*Miscanthus*[b]			
		AD	GA	CO	CF	AD	GA	CO	CF	AD	GA	CO	CF
Cultivation	Yield (t DM ha^{-1} y^{-1})	14				13				10			
	Yield (t FM ha^{-1} y^{-1})	77				25				11			
	$Energy_{db}$ (GJ ha^{-1} y^{-1})	230				230				180			
	$Energy_{wb}$ (GJ ha^{-1} y^{-1})	77				200				180			
Pre-treatment & Storage	El. (MWh ha^{-1} y^{-1})	0.10	0.10	-	0.10	0.096	0.096	-	0.096	0.074	0.074	-	0.074
	Heat (GJ ha^{-1} y^{-1})	-	-	-	-	16	-	-	-	13	-	-	-
	DM loss (t DM ha^{-1} y^{-1})	3.3				0.61				0.55			
Operation	El. (MWh ha^{-1} y^{-1})	0.78	0.34	4.6[c]	4.6[c]	0.89	0.31	5.1[c]	5.1[c]	0.72	0.74	6.1[c]	6.1[c]
	Heat (GJ ha^{-1} y^{-1})	9.3	-	-	-	12	-	-	-	9.4	-	-	-
Crop fed	Crop fed (t DM ha^{-1} y^{-1})	10				12				9.4			
	Crop fed (t FM ha^{-1} y^{-1})	12				14				11			
	$Energy_{db}$ (GJ ha^{-1} y^{-1})	170				220				170			
	$Energy_{wb}$ (GJ ha^{-1} y^{-1})	170				210				170			
Raw pig manure	Amount (t DM ha^{-1} y^{-1})	4.7	-	-	-	6.3	-	-	-	5.0	-	-	-
	Amount (t FM ha^{-1} y^{-1})	69	-	-	-	92	-	-	-	72	-	-	-
Gas conversion	$Energy_{gas}$ (GJ ha^{-1} y^{-1})	140	120	-	-	160	150	-	-	130	120	-	-
Energy efficiency	η_{el} (%)	38	38	27	38	38	38	27	38	38	38	27	38
	η_{ht} (%)	52	52	63	52	52	52	63	52	52	52	63	52
Net energy output	El. (MWh ha^{-1} y^{-1})	14	13	13	18	16	16	16	23	13	12	13	18
	Heat (GJ ha^{-1} y^{-1})	65	64	110	88	56	80	140	110	45	62	110	88
Overall energy conversion from crop	$\eta_{tot\ el}$ (%)	22	20	20	28	26	25	25	36	26	25	26	36
	$\eta_{tot\ ht}$ (%)	28	28	47	38	24	35	61	48	25	35	61	49

[a] FM: Fresh matter; DM: dry matter; db: dry basis; wb: wet basis; η_{el}: electricity efficiency; η_{ht}: heat efficiency; η_{tot}: total efficiency
[b] AD: anaerobic digestion; GA: gasification; CO: Combustion; CF: Co-firing
[c] The electricity consumption is reported although this is already accounted for in the net efficiency reported in the line 'Energy efficiency'.

9.5 Mass flow analysis of C and N

Carbon and nitrogen flows are two of the most important flows responsible for the environmental impacts involved in bioenergy systems. Therefore, the C and N flows of all the scenarios assessed in this case study have been disaggregated and calculated for all the major processes involved. This is illustrated in Figures S13-S18 of Tonini *et al.* (***II***) (SI). One example for the C flows involved in the case of anaerobic co-digestion of pig manure with *Miscanthus* (spring harvested) is presented in Figure 16.

As illustrated in Figure 16, only ca. 40% of the C uptake from the atmosphere ends up in the harvested biomass, the rest ending up in the non-harvested above- and below-ground residues (as also shown in Table 18[71]). Further, it should be highlighted that more than 85% of the C input to the energy crop system ends up emitted as CO_2, whether as a result of the cultivation stage or as a result of the final energy use. Figure 16 also illustrates that most of the digestate' C ends up emitted as CO_2 rather than entering the soil C pool.

As opposed to C, the outputs of N flows were more diversified among the individual flows. The most significant N flows occurred during the land application of the digestate for the anaerobic co-digestion scenarios, and during the cultivation stage for the other scenarios (Figures S16-S18 and Table S8 of the SI of Tonini *et al.* ***II***). Overall, NO_3^- and NH_3 emissions were the most significant N-emissions.

[71] However, Table 18 presented values on sandy soil, while the present case study considered a sandy loam soil.

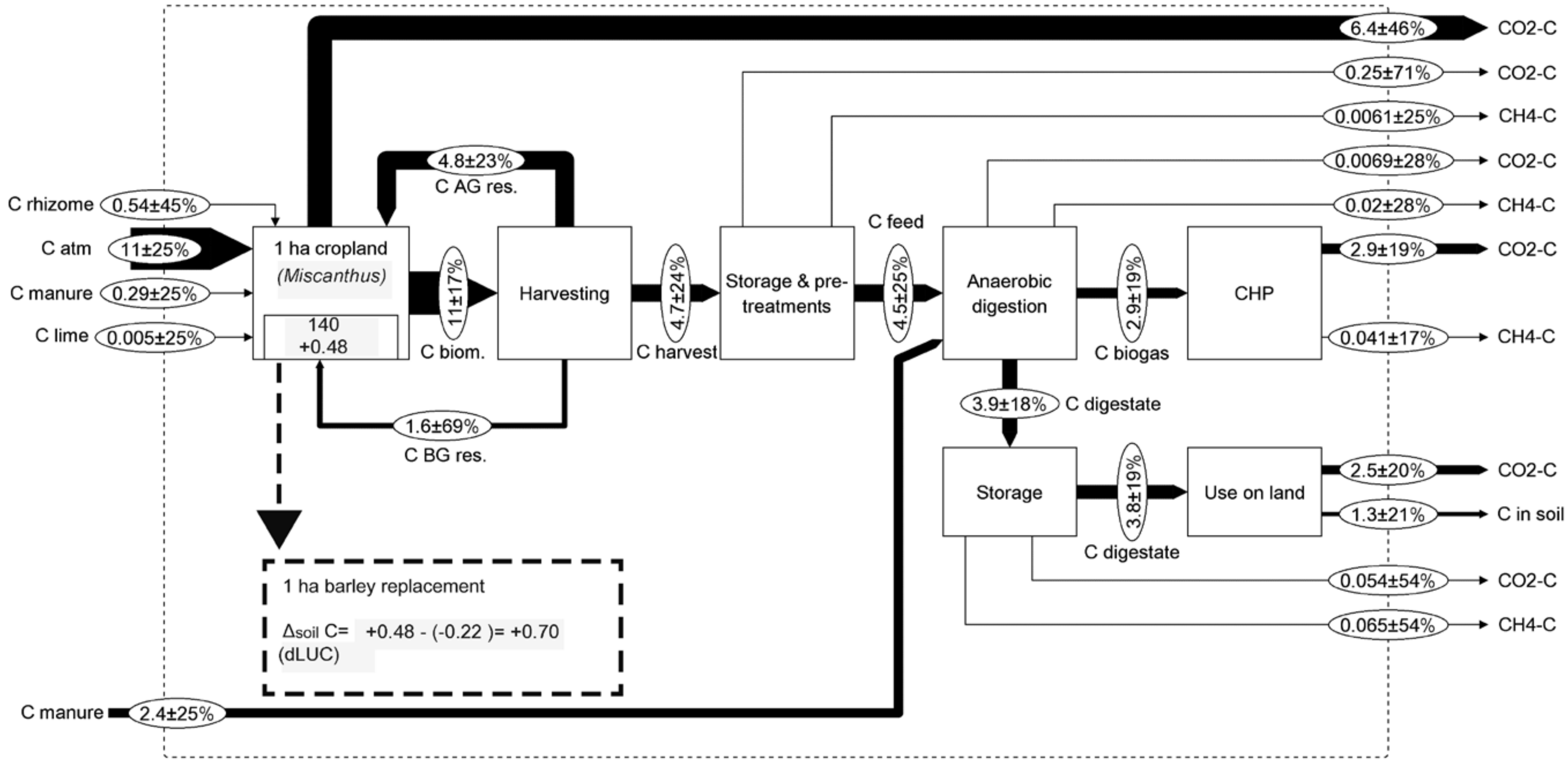

Figure 16. Illustration of the C flows breakdown (t C ha^{-1} y^{-1}) for anaerobic co-digestion of *Miscanthus* with pig manure (values rounded to two significant digits)

9.6 LCA results

The LCA results of this case study are illustrated in Figure 2 of Tonini *et al.* (***II***), with breakdown per process, while the net results for the environmental impact categories addressed are summarized in Table 27.

Table 27. Overview of the LCA results for the bioenergy scenarios assessed in Tonini *et al.* (***II***)[a]

Process	**Ryegrass**				**Willow**				***Miscanthus***			
	AD	**GA**	**CO**	**CF**	**AD**	**GA**	**CO**	**CF**	**AD**	**GA**	**CO**	**CF**
Global Warming (kg CO_2 eq. ha^{-1} of perennial crop)												
DLUC	-167	-167	-167	-167	-249	-249	-249	-249	-211	-211	-211	-211
ILUC	309	309	309	309	309	309	309	309	309	309	309	309
Crop pre-treatment	118	118	118	118	46	28	28	28	42	24	24	27
Energy production	285	343	351	351	316	426	440	440	252	322	336	335
Energy substitution	-375	-339	-416	-482	-448	-433	-521	-612	-355	-330	-432	-504
Use on land, net[b]	145	0	0	0	139	0	0	0	97	0	0	0
Raw manure storage	-84	0	0	0	-112	0	0	0	-88	0	0	0
Other	20	1.6	1.7	1.7	56	1.8	1.9	1.8	50	0.6	1.6	-0.83
Net[c]	**251**	**266**	**197**	**131**	**57**	**83**	**9**	**-82**	**97**	**115**	**28**	**-45**
Aquatic Eutrophication (N) (kg N eq. ha^{-1} of perennial crop)												
DLUC	434	434	434	434	-569	-569	-569	-569	-550	-550	-550	-550
ILUC	0	0	0	0	0	0	0	0	0	0	0	0
Crop pre-treatment	3	0	0	0	2	0.08	0.08	0.08	2	0.06	0.06	0.06
Energy production	55	43	41	41	63	52	34	34	50	41	54	43
Energy substitution	-32	-29	-34	-41	-38	-36	-43	-52	-29	-28	-35	-43
Use on land, net[b]	1820	0	0	0	580	0	0	0	360	0	0	0
Raw manure storage	-49	0	0	0	-66	0	0	0	-51	0	0	0
Other	39	4	3	4	48	0.9	0.9	1	36	0.9	1	0.5
Net[c]	**2270**	**452**	**444**	**438**	**20**	**-552**	**-577**	**-586**	**-182**	**-536**	**-530**	**-550**
Aquatic Eutrophication (P) (kg P eq. ha^{-1} of perennial crop)												
DLUC	-10	-10	-10	-10	-2	-2	-2	-2	-3	-3	-3	-3
ILUC	0	0	0	0	0	0	0	0	0	0	0	0
Crop pre-treatment	0	0	0	0	0	0	0	0	0	0.07	0	0
Energy production	0.01	0.4	0.02	0.03	0.01	0.3	0.03	0.03	0.03	0.3	0.03	0.03
Energy substitution	-0.2	-0.2	-0.3	-0.4	-0.2	-0.3	-0.4	-0.4	-0.2	-0.2	-0.3	-0.4
Use on land, net[b]	53	0	0	0	16	0	0	0	49	0	0	0
Raw manure storage	0	0	0	0	0	0	0	0	0	0	0	0
Other	1	0.1	0.1	0.2	1	0.1	0.1	0.1	2	-4	-3	-3
Net[c]	**44**	**-9**	**-10**	**-10**	**15**	**-2**	**-2**	**-2**	**49**	**-6**	**-7**	**-7**
P as a resource (kg P ha^{-1} of perennial crop)												
DLUC	-157	-157	-157	-157	48	48	48	48	0	0	0	0
ILUC	0	0	0	0	0	0	0	0	0	0	0	0
Crop pre-treatment	0	0	0	0	0	0	0	0	0	0	0	0
Energy production	0	0	0	0	0	0	0	0	0	0	0	0
Energy substitution	-0.1	-0.2	-0.4	-0.4	-0.2	-0.2	-0.4	-0.4	-0.2	-0.2	-0.5	-0.3
Use on land, net[b]	0	0	0	0	0	0	0	0	0	0	0	0
Raw manure storage	0	0	0	0	0	0	0	0	0	0	0	0
Other	25	-1	0	0	26	-1	-1	-1	36	-0.1	0.01	-0.2
Net[c]	**-132**	**-158**	**-158**	**-158**	**74**	**47**	**47**	**47**	**36**	**-0.2**	**-0.4**	**-0.4**

[a] AD: Anaerobic digestion; GA: Gasification; CO: Combustion; CF: Co-firing.
[b] Digestate minus raw manure
[c] Eventual inconsistencies due to rounding.

9.7 Sensitivity and uncertainty analyses

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

Parameter uncertainties were addressed through a MonteCarlo analysis (number of simulations: 1000), whereas scenario uncertainties were addressed through sensitivity analyses. These included: a) variation (min-max) of the iLUC impacts with respect to CO_2 emissions (vs. mean value assumed as baseline); b) winter wheat as the marginal crop for Denmark (vs. spring barley as baseline); c) coal-based heat production as the marginal technology for heat generation (vs. natural gas-based as baseline); d) natural gas power plant as the marginal technology for electricity generation (vs. condensing coal power plant as baseline); e) mono-digestion of the crops (vs. co-digestion with manure as baseline); f) pre-treatment of pelletization before co-firing (vs. 'no pelletization' as baseline). Each of these changes was tested individually to assess the influence of the individual change on the overall LCA results. The sensitivity analyses results are presented in Figure S19 of Tonini *et al.* (***II***) (SI), and the Monte Carlo results in Table S18 of Tonini *et al.* (***II***) (SI).

9.8 Overview of main findings

The main findings of this case study can be summarized as follows:

- Only co-firing of willow and *Miscanthus* indicated net overall global warming savings, i.e. these were the only two scenarios for which an environmental benefit, GHG-wise, was identified in relation to using 1 ha of land for bioenergy.
- The market-driven land expansion (i.e. indirect land use change) resulting from using more Danish arable land for energy crop cultivation was shown to offset any potential benefits of bioenergy, except for *Miscanthus* and willow co-firing.
- None of the assessed scenarios could achieve a GHG reduction of 35% in comparison with a reference fossil-fuel system.
- Cultivating *Miscanthus* and willow instead of spring barley did result in an environmental benefit for global warming and aquatic eutrophication (N & P), on the perspective of the cultivation system only.
- The results of the sensitivity analyses highlighted that the variation of the ILUC impact played the most important role for global warming; with minimum ILUC impacts (7 t CO_2 eq. $ha^{-1}y^{-1}$) all bioenergy scenarios for willow and *Miscanthus* as well as co-firing of ryegrass achieved environmental savings. In all other analyses, the individual changes in assumptions did not alter the conclusions relative to the baseline, although the net GWPs were affected. The results of the MonteCarlo simulation supported the ranking of the bioenergy scenarios found with the baseline scenarios, except between *Miscanthus* and willow thermal conversions (gasification, combustion and co-firing), where it appeared unclear if willow was really better than *Miscanthus*.

10. DISCUSSION AND PERSPECTIVES

Studies available to date indicate that a Danish renewable energy system would require between 300 and 450 PJ y^{-1} of biomass, which represents ca. 35-50% of today's overall energy consumption (Lund *et al.* 2011; Danish Commission on Climate Change Policy 2010; Energinet.dk 2010). As mentioned in section 1, there are three main reasons explaining why such an important share of biomass is necessary in a fossil-free energy system:

i. It can be used for producing high energy density fuels needed for the portion of the transport sector that cannot be electrified (aviation, long-distance road and sea transport);
ii. It can be used to synthetize C-based chemicals and materials;
iii. It can be stored, and as such can be used to ensure flexibility in balancing the fluctuating energy production from intermittent sources like wind, solar and wave power.

These three main "customers" for biomass all have the same common denominator: carbon. In a renewable energy system, biomass in fact represents the main supplier of C. Yet, this PhD work emphasized that biomass, which is also solicited by increasing demands from the projected population growth, dietary changes, and bioenergy policies worldwide, is constrained. Consequently, the C available in a renewable energy future is correspondingly limited.

In this perspective, this constrained C should, in a renewable energy system, be used as efficiently as possible. This involves 2 main prioritization aspects:

- Prioritizing the biomass-to-energy (BtE) conversion pathways allowing the greatest efficiency in fulfilling the demands from the three main biomass customers mentioned above;
- Establishing a priority order with respect to the types of biomass to use in a Danish renewable energy system.

These prioritization issues represent the key implications of the findings made within this PhD work.

10.1 Prioritizing the biomass-to-energy conversion pathways

The future Danish renewable energy system will rely primarily on wind power for electricity production. This is well illustrated with the Danish Government goal to have half of its electricity consumption supplied by wind already in 2020 (Danish Government 2011) and by various studies analyzing future designs of a 100 % renewable energy system in Denmark (Lund *et al.* 2011; Danish Commission on Climate Change Policy 2010; Energinet.dk 2010), all basing the design on a much higher wind power to total electricity ratio, above 75% in most cases. Such a system involves that the limited available residual biomass must, in order to avoid/minimize the need for land-dependent energy crops, be used mainly for regulating power, i.e. in the periods where wind power cannot supply the full electricity demand, as illustrated in Figure 17.

In Figure 17, two key aspects of an electricity system involving a high share of fluctuating wind power are illustrated:

a. Periods of electricity surplus, where the power produced from wind mills exceeds the electricity demand (green areas above the demand curve);
b. Periods of electricity deficit, where wind power is not sufficient to cover the electricity demand (pink areas).

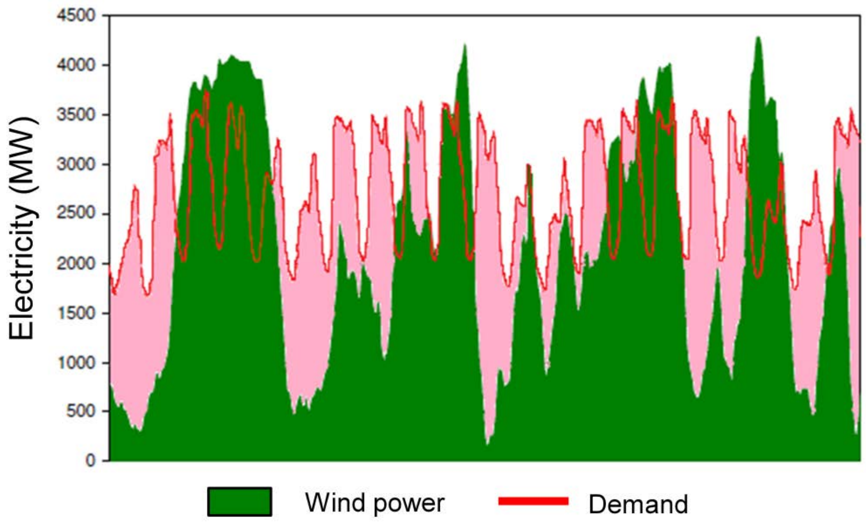

Figure 17. Electricity produced from wind power versus electricity demand, forecasted for 2020 (modeled as 2008's wind capacity + 3000 MW) and illustrated for the month of January, i.e. for the 744 hours of the month (X axis). The pink area represents the periods where the demand exceeds the wind production. Adapted from Hansen (2011).

Efficient use of the biomass for electricity balancing, or rather its quantity as a C-containing storable fuel, implies that biomass should be used only when wind power is not sufficient to cover the electricity demand (pink areas of Figure 17). Any BtE conversion pathway involving that biomass is competing with wind (green areas under the demand curve in Figure 17) can thus be seen as "C inefficient", and is obviously not a desirable situation, neither on an economic nor environmental perspective. This is among other the case for combustion in CHP biomass plants, as the electricity generated from these plants must be used immediately after it is produced. In this perspective, this is where "bio"-gas[72] (whether it is obtained through thermal gasification or anaerobic digestion technologies) becomes a key advantage in a renewable energy system: being storable in the natural gas grid, it allows using the biomass exactly when it matters the most (pink areas in Figure 17).

"Bio"-gas thus offers a major efficiency advantage with respect to the third of the three above-mentioned priority customers for biomass, i.e. balancing the fluctuating energy production from intermittent resources. Furthermore, "bio"-gas, as stored in the gas network, can concomitantly be used for supplying the demands from the transport sector (the first of the three priority customers), both for air, sea and road (long-distance), which represents by far the greatest demand for biomass in a renewable energy system (Lund *et al.* 2011). In fact, "bio"-gas can be whether compressed and used directly, or used to synthetize so-called syn-fuels (e.g. dimethyl ester or methanol). Similarly, the stored "bio"-gas can be used to synthetize C-based chemicals and materials (second priority customer). "Bio"-gas would thus allow, in this way, to integrate the transport and material/chemical sectors into the energy system, thereby allowing to limit the unnecessary use of biomass that could result from a poor coordination between these different biomass (or C) demanding sectors. On this sense, it is "C efficient" solutions that should be favored rather than "energy efficient" solutions: in fact, it is C that is constrained, while energy as such is not.

The technologies allowing for the lowest global warming potentials were, in Tonini *et al.* (***II***), found to be the state-of-the-art combustion technologies, which were also the technologies allowing the greatest overall energy efficiencies (Table 26). These, as earlier described, however involve an inevitable competition with wind power. This highlights that, technology-wise, one important challenge for reducing the overall GHG

[72] This expression is employed instead of "biogas" in order to avoid confusion: biogas is indeed typically used to refer to the gas obtained from anaerobic digestion.

emissions of future renewable energy systems lies in increasing the C conversion efficiency of biomass in anaerobic digestion and gasification technologies. One way to do this could be to thermally gasify the digestate obtained from anaerobic digestion (instead of applying it on land), where a maximum of the biomass C could then be converted to energy, just like in the incineration case (which would, on the other hand, imply a certain nutrient loss, and return less C to soil). Another way could be through hydromethanation, where the CO_2 portion of biogas, instead of being stripped off prior to biogas injection into the gas grid, is converted to additional CH_4 through a reaction with hydrogen (Wenzel 2010; Sterner & Fritsche 2011).

Transport has, in general, not been included in this PhD work, and as a result, no comparison between the above-described syn-fuels (which can be generated from "bio"-gas directly, from "bio"-gas combined with H_2[73], or from combining H_2 to the recycled CO_2 captured from combustion processes) and biofuels (whether from energy crops, biowastes[74], straw or other lingo-cellulosic residues) were made. However, as shown in Wenzel (2010), the energy conversion efficiency, i.e. the amount of energy that can be produced from the initial energy content of the biomass, is rather poor for most biofuels, as compared to the possibilities offered by syn-fuels. For example, Wenzel (2010) showed that for 100 PJ energy in the biomass, only 70 PJ (including 20 PJ in the co-product) are created with biofuels produced through typical fermentation pathways, while between 170 and 340 PJ can be produced through syn-fuel pathways[75], from the same initial 100 PJ of biomass. On this perspective, it appears that these fuels offer greater possibilities to reduce the amount of biomass needed for a renewable energy system. It can be argued that some biofuels (depending on the feedstock) also allow to recover the protein portion of the feedstock, thereby releasing pressure on land. Assessing whether this "transport strategy" to produce fuels and reduce land use changes yields greater benefits than reducing the demand of land-dependent biomasses through producing syn-fuels was however beyond the scope of this study.

10.2 Agricultural biomass prioritization

The case studies performed in Hamelin/Tonini *et al.* (***II, IV, V***) all highlighted the tremendous GHG reduction potential involved in avoiding, through biogas production, the reference manure management to take place (i.e. when conventionally storing raw manure and applying it on land, without further processing). Recent studies also presented similar findings (e.g. Sterner & Fritsche 2011; Meyer-Aurich *et al.* 2012; De Vries *et al.* 2012). This is mostly due to avoiding the releases of CH_4 to atmosphere during raw manure storage, a greenhouse gas with a global warming potential 25 times the one of CO_2, on a 100 years horizon (IPCC 2007). When using manure for biogas production, this CH_4 is instead used to generate fossil-free energy (thereby displacing the use of marginal energy, and the GHG emissions related to it), after which it is emitted to atmosphere mostly as CO_2, instead of being emitted as CH_4 as in the reference manure management case. Although digested manure may, in comparison to raw manure, contribute to additional ammonia emissions during storage and spreading (Sommer *et al.* 2006; Moeller & Stinner 2009), this drawback can be mitigated through, for example, acidification of the digestate (e.g. Wesnæs *et al.* 2009). Other benefits of manure digestion (with land application of the digestate), include a reduced potential for nitrate losses (Sørensen & Birkmose 2002) and an increased N efficiency of the fertilizer (i.e. more N is in an inorganic form, which plants can use; Jørgensen 2004), among others.

In a prioritization perspective, this highlights that, in a renewable energy system, manure should be prioritized for biogas. This allows for two major benefits: 1) reducing the overall GHG emissions from the manure management sector and 2) allowing for the production of a storable gas, enabling both intermittent power production, C-based materials and chemicals production and the production of transport fuels.

[73] In this case, the H_2 would be produced through water electrolysis, and the electricity input to the process would be provided by the surplus wind, as detailed in Wenzel (2010).

[74] Biowaste is not an agricultural biomass as such, but is mentioned here as it was investigated in Hamelin *et al.* (***V***), where it consisted of food waste (whether from households and commercials) as well as garden waste.

[75] This involves the inclusion of H_2 in hydrocarbon synthesis through hydrogenation.

Besides the reference manure management system, manure could also be processed for energy via combustion. Such mass burning, as earlier detailed, not only presents the disadvantage to compete with wind power, but would further results into little environmental benefits (in comparison to the reference manure management), due to the energy needed to evaporate the water content of the manure. This was shown in Hamelin *et al.* (***IV***) (alternative P3) for the case of pig slurry. The energy balance may be slightly different if, for example, deep litter is burned, but again the flexibility benefit offered by biogas is then lost, as well as a significant share of the nutrients and C.

It was also highlighted in Hamelin *et al.* (***IV, V***), that manure-based biogas needs to be supplemented by C-containing co-substrates in order to ensure the economic sustainability of biogas production, or in other words, to ensure that a wide-spread implementation of manure digestion can happen. Among all co-substrate alternatives investigated, source-segregated solid manure was the option yielding, by far, the greatest environmental benefits, particularly because it allowed significantly more manure to be diverted from the reference manure management towards biogas production. However, this option involves significant changes to the animal houses already in place throughout Denmark (in terms of infrastructure), and consequently important investments. Until such changes/investments take place, others options to increase the amount of CH_4 produced from manure-based biogas in the short- to medium-term future must be considered.

In this perspective, and in the light of Denmark's ambition to digest 50% of its manure by 2020, straw and biowastes (whether it is garden waste or food waste from households and commercials) were highlighted as co-substrates to prioritize for co-digestion with manure. This is because their use for co-digestion with manure:

- Resulted in a lower global warming potential (compared to their other uses), as shown in Hamelin *et al.* (***V***);
- Prevented energy crops to be used for co-digestion[76];
- Allowed the nutrients of these biomasses to be recycled, their degradable C content to be used for energy production, and their non-degradable C content to enhance the content of soil organic C;
- Allowed the production of a storable and versatile gas.

In a nutshell, it is concluded that the residual biomasses studied within this PhD work, i.e. food waste (household and commercial), garden waste and straw, should, in the perspective of a renewable energy system, be prioritized for the production of a storable biogas through co-digestion with manure. This conclusion, however, should not be seen as universal and applying for all organic residues. In fact, if a residue (e.g. beet tops) have a protein value, using it for feed is likely to translate into greater environmental benefits than using it for biogas, in particular because of the land use changes it then avoids. This was among others demonstrated in De Vries *et al.* (2012).

It should further be noted that additional co-substrates would deserve to be considered in an analysis like the one performed in Hamelin *et al*. (***V***). Particularly, the co-digestion of deep-litter with raw manure/slurry could represent an important potential for increased methane production. In fact, a recent study by Birkmose *et al.* (2013) estimated the 2020 potential of deep litter in Denmark to represent ca. 185 million Nm^3 CH_4 y^{-1}, being the third largest available potential for CH_4 production after straw (600 - 740 million Nm^3 CH_4 y^{-1}) and slurry (335 million Nm^3 CH_4 y^{-1}) itself. In future work, an additional scenario including co-digestion with deep litter should thus be investigated.

The case studies performed within this PhD work (Hamelin *et al.* ***V***; Tonini *et al.* ***II***) highlighted the potentially tremendous environmental impacts of using energy crops within a renewable energy system, because of the

[76] In a LCA comparing e.g. co-digestion and incineration of biowastes, in the context of the Danish ambition to digest 50% of the manure by 2020, this should be seen as the "lost alternative".

land use changes (direct and indirect) they generate. Particularly, it was shown that the cascading effects induced by using more land for energy crops within a renewable energy strategy in Denmark was, in most bioenergy scenarios assessed, leading to an increase of GHG emitted to the atmosphere in comparison to the GHG offset by the crops substituting fossil fuels.

In this perspective, the use of energy crops should be at the bottom of the "priority order" for bioenergy production in a renewable energy strategy. However, as mentioned in section 1, it is not realistic to completely avoid energy crops in a renewable energy system. There are nevertheless ways to minimize the impacts of energy crops.

For example, Hamelin *et al.* (***I***) showed that some crops allowed the attainment of much greater yields than other crops (Table 17), while enhancing the SOC content (Figure 9) and allowing for lower nutrient losses (Figure 12). These crops consist of the perennial crops *Miscanthus* and willow, and their environmental benefits are summarized in Table 28, in comparison to maize silage.

Table 28. Environmental benefits of perennial crops, illustrated in comparison to maize silage

	Soil C changes after 20 years[a] (kg C ha^{-1})	Yield (t DM ha^{-1} y^{-1})	DM (% FM)	Net GWP_{100}[d] (t CO_2 eq. ha^{-1} y^{-1})	Net eutrophication-N potential[d] (kg N eq. ha^{-1} y^{-1})	Net eutrophication-P potential[d] (kg P eq. ha^{-1} y^{-1})
Miscanthus-A[b]	-660 to 3660	13 to 15	44%	-18.5	13.9	0.20
Miscanthus–S[c]	8410 to 12180	8.5 to 10	85-90%	-12.6	13.2	0.30
Willow	-1860 to 10690	7.1 to 13	50%	-13.6	12.4	0.54
Maize silage	-10190 to -4370	12.0	31%	-13.5	60.9	1.38

[a] A negative value indicates a loss of native soil C, whereas a positive value indicate an increase of soil organic C. The wide range reflects the different *soil×climate* combinations. For willow, it also includes the values for the year where fertilization is performed with 100% mineral fertilizers (a negative value is actually only found for willow cultivated on a sandy soil, under a "dry" climate and with 100% mineral fertilizers).
[b] Autumn harvest
[c] Spring harvest
[d] For a sandy soil, under a wet climate (964 mm y^{-1}). These values are for the cultivation stage only; storage and final fate of the crop are not included.

As it can be seen from Table 28, eutrophication results (N and P) are very similar for *Miscanthus* (both harvest seasons) and willow, and clearly lower than for silage maize. All crops present relatively high yields (in comparison to e.g. spring barley), but autumn harvested *Miscanthus* appears to be the crop allowing the highest yields. However, this crop presents the drawback of native soil C losses, if cultivated on a sandy loam soil. On the other hand, spring harvested *Miscanthus* has a slightly lower yield, but allows for increases in soil C for all *soil×climate* combinations assessed in Hamelin *et al.* (***I***). Further, it has the advantage (in comparison to autumn harvested *Miscanthus*, but also to all other crops assessed) to have a very high DM content at harvest (i.e. a low water content). A high DM content is indeed desirable, and required, for thermal conversion processes. For example, the gasification process in fluidized bed typically requires biomass with water content below 20% (Hughes & Larson 1998). Although some combustion plants are able to burn woody biomass (including willow) with 50% moisture, such high water content is not desirable as it tends to create problems in the boiler, among other because of the increased flue gas then produced[77]. As emphasized in Tonini *et al.* (***II***), one simple way to handle high moisture content consists to dry the willow rods or mowed *Miscanthus* on-field, although this is associated to potentially considerable DM losses, particularly for *Miscanthus*, being a grass specie. Albeit counter-intuitive, dryer biomasses are also desirable for anaerobic co-digestion with animal slurries. In fact, biogas plants are interested, within technical limits

[77] Personal communication with Niels Ole Knudsen, Dong Energy, November 2011.

(section 8.2), to incorporate as much DM as possible in the input mixture in order to produce greater amounts of energy (this applies, of course, in the perspective of co-digestion with animal slurries, these presenting a very low DM content).

Spring harvested *Miscanthus*, under this perspective, therefore appears as an energy crop to favor for a renewable energy strategy, although energy crops should be at the bottom of the priority order. Of course, it would not be advisable to rely on only one crop to provide the “energy crop portion” of the biomass needed for a renewable energy system, especially in the light of reducing the biomass vulnerability to eventual pest outbreaks. In this perspective, willow also represents an interesting crop to consider.

10.3 Perspectives for integration with the waste sector

Hamelin *et al.* (***V***) showed that ca. 90% of the household biowaste produced in Denmark is incinerated today (SI, Table S59). Yet, as highlighted above, there are many advantages to use this food waste for co-digestion with manure instead of incinerating it together with the residual waste in CHP plants. Besides an improvement of the GHG, nutrients, or soil C balance, anaerobic co-digestion allows the production of a storable gas, while incineration involves a competition with wind.

Of course, the fate of wastes like household biowaste is today determined in a waste management perspective. Diverting food waste towards anaerobic co-digestion would, thus, induce interactions with the waste sector. For example, how best should this biowaste fraction be separated from the residual waste? How would this affect the incinerators performance, and would this induce an import of waste from abroad to compensate for the lost waste? What would be the consequences of this? Answering such questions was obviously beyond the scope of this PhD work, but these emphasize the necessity to integrate the waste sector consistently into a renewable energy strategy.

10.4 Key uncertainties

10.4.1 Straw harvest

One major uncertainty regards the impact that the use of straw for bioenergy would have on soils quality. This was already partly discussed in section 3.2. In the perspective of a renewable energy system, it would, thus, be convenient to fix a criteria, e.g. a universal SOM threshold, below which straw should not be harvested in order to preserve/improve soils quality.

It is however well acknowledged that there is no universal SOM threshold with regards to soils capacity to support sustainable tilth conditions across all soil types (Schjønning *et al.*, 2009). As determining the critical SOM level to maintain at the local/micro level all over Denmark/Europe would represent a rather tedious task, Schjønning *et al.* (2009) proposed to use another indicator to quantify soil’s vulnerability, i.e. the clay: SOM ratio, which they refer to as the “Dexter ratio”, since based on the study of Dexter *et al.* (2008). According to this, any soils above a Dexter ratio of 10 can be considered as vulnerable. Schjønning *et al.* (2009) represented the vulnerability of Danish soils, expressed in terms of the Dexter ratio, for the whole of Denmark, where the soils above a Dexter ratio of 10 can be visualized (consisting of most areas of Lolland and Bornholm, among others)[78]. Most of these vulnerable areas coincide with the areas where soils are richer in clay. These soils were also pointed out, in Hamelin *et al.* (***I***), as those where the highest soil C losses occurred.

In an endeavor to sustainably integrate the agricultural sector into a renewable energy strategy, it would thus be advisable to increase SOM in these high Dexter ratio areas, whether through avoiding the harvest of residues, or through favoring the cultivation of crops leading to the greatest increases in SOC (willow, spring harvested *Miscanthus*, ryegrass or winter wheat with straw incorporation), providing these crops have to be cultivated anyway. Further, the yield of these crops is slightly higher on soils richer in clay, as shown in

[78] Figure 6.16 of Schjønning *et al.* (2009).

Hamelin *et al.* (***I***). On the other hand, straw harvest could be favored in the areas with low Dexter ratio, which is the case for most of Jutland, and significant portions of Funen and Zealand. Similarly, low Dexter ratio areas should be prioritized for cultivating crops leading to soil C decreases (e.g. barley, maize silage or sugar beet), to the extent these have to be cultivated.

It should also be highlighted that straw harvest does not necessarily implies a loss of soil C. In fact, if straw is used e.g. for co-digestion with animal slurry and the digestate then produced is returned to land as a fertilizer, little soil C will be lost. Of course, the digested material returns less C to the soil in comparison to the overall C content of the biomass prior to digestion, as the easily degradable C of it is diverted to the biogas. On the other hand, if applied directly on land (i.e. without digestion), most of this easily degradable C would not contribute to enhance the soil C pool either, but would simply be emitted as CO_2. This was actually quantified in Hamelin *et al.* (***IV***), for the case of pig slurry, and it was shown that between 13 to 38[79]% less carbon ended up in the soil C pool with the different digested slurries assessed, as opposed to raw slurry. Considering that straw removal (without any return) would, for example, decrease the soil C by 390% in the spring barley cultivation system (Hamelin *et al.* ***I***)[80], this suggests that the soil C losses due to anaerobic digestion are of minor importance.

According to recent studies (Li *et al.* 2005; Cherubini *et al.* 2009; Börjesson & Tufvesson 2011), straw incorporation, although it allows to increase soil C, could also lead to greater N_2O emissions, because of 1) of the increased N input to soil and 2) the concomitant increased C input to soil, which would spur the activity of the soil biota and as such enhance nitrification. In this PhD work, only the first aspect was taken into account, for the reason explained in footnote 51. However, it resulted to a minor effect: i.e. N_2O increases of ca. 3% for winter wheat straw, 8% for spring barley straw, but 19% for sugar beet tops. In the light of these results, it does not appear that a strategy favoring e.g. the cultivation of crops giving rise to important above-ground biomass would lead to an increase of N_2O emissions hindering the GHG balance of such strategy. Further, Mutegi *et al.* (2010) recently showed, in a Danish field experiment, that crop residues input does not necessarily cause higher N_2O emissions. Nevertheless, at the light of establishing a sustainable bioenergy strategy for Denmark, the impact of above-ground residues on N_2O emissions should, ideally, be investigated through field trials for a combination of Danish soils and cultivation systems.

10.4.2 DLUC database

The DLUC database established in this study (Hamelin *et al.* ***I***) covers crops that would likely be affected by a renewable energy strategy in Denmark (whether through an increase or decrease). To enhance the significance of the database, one additional crop, namely rapeseed, could be included. Although rapeseed is a crop involving high pesticides input as well as a relatively low yield, it is likely to be considered as a crop to be used for fulfilling the obligations Denmark has in the framework of the Renewable Energy Directive (at least 10% of the final energy from the transport sector should be supplied by renewables, by 2020). As such, including rapeseed into the DLUC database would likely represents a useful tool for decision makers wanting to address the environmental impacts of this crop in a LCA perspective. Similarly, as highlighted in Tonini *et al.* (***II***), lower N application could be considered for ryegrass (in the perspective that it is dedicated to bioenergy), where protein production is then not the focus (as in the case of forage ryegrass).

Further, the DLUC database considered that crops' N needs were fulfilled through 50% mineral fertilizers and 50% manure. Through this had the benefit of reflecting the implication of manure fertilization on various substances flows (e.g. NH_3, N_2O, P and N losses) as well on soil C changes, it does not fully follow the consequential LCA principles, according to which it is the marginal suppliers that would react to an increased demand for these crops (and thus of the fertilizers for these crops). On this sense, the inventory should ideally also be made with 100% mineral fertilizers.

[79] This is for the 2 scenarios were no C is lost through combustion, i.e. alternatives P1 and P2 (part of the manure pellets produced in alternative P3 were in fact burned to produce to energy required for the drying process)
[80] On sandy soils. For winter wheat (also on sandy soil), the decreased would be 80%.

10.4.3 N_2O from field processes

According to Nieder & Benbi (2008), N losses by denitrification of managed and unmanaged ecosystems are the biggest unknown in the terrestrial N cycle. In biosystem LCAs, uncertainties related to N_2O are often pointed out as having significant effects on the outcome of the results (i.e. high sensitivity) (e.g. Cherubini *et al.* 2009; Meyer-Aurich *et al.* 2012). This among others reflects the important GWP of N_2O, being 298 times the one of CO_2 (IPCC, 2007; for a 100 years horizon).

Most of the N_2O in crop systems occurs through microbiological transformation of N (Oenema *et al.* 2005) and this involves three main processes: nitrification, denitrification and nitrifier denitrification[81]. Figure 18, adapted from Oenema *et al.* (2005), illustrates the microbiological processes leading to N_2O emissions.

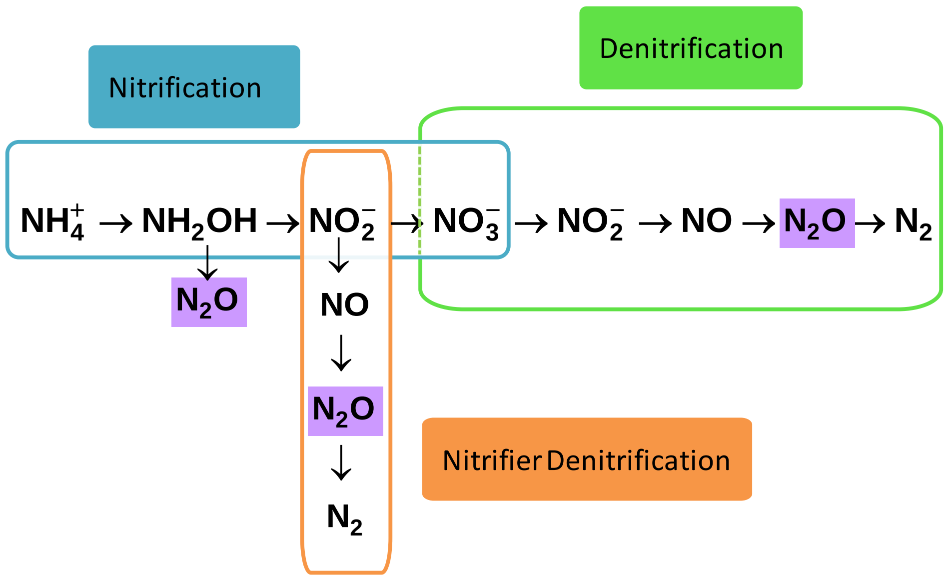

Figure 18. Illustration of the biological processes leading to N_2O emissions, adapted from Oenema *et al.* (2005)

As shown in Figure 18, nitrification is a pre-condition for N_2O to occur, and this requires aerobic conditions. On the other hand, anaerobic conditions are necessary for denitrification to occur[82]. Under partial or transient anaerobic conditions, the denitrification reaction is uncompleted, resulting in the production of NO and N_2O.

Thomsen *et al.* (2010) proposed that the propensity of soils to emit N_2O instead of N_2 can be represented as a bell-shape curve (Gaussian distribution), with the ratio O_2 demand to O_2 supply on the X axis, and the emissions of N_2O (and N_2) on the Y axis. According to Thomsen *et al.*'s theory, applying e.g. digested slurry (which represents a reduction in O_2 demand compared to raw slurry) would contribute to decrease N_2O emissions on dry soils (and increase N_2; Figure 17), but to increase them on wet soils. Similarly, injecting slurry (which represents a reduction in O_2 supply compared to raw slurry) would contribute to increases of N_2O emissions on dry soils, but decreases on wet soils. This illustrates the importance of site-specific conditions with respect to estimating N_2O emissions.

In the framework of this PhD work, a proper estimation of the N_2O emissions is important in particular in the perspective of estimating the impacts of intensification. According to the simulations made by Melillo *et al.* (2009) up to 2100, N_2O emissions due to increased fertilization (occurring as a response to an increased

[81] In addition to these microbiological processes, N_2O may also be formed chemically through chemodenitrification, but this is generally not considered as an important source of N_2O (Yates 2006).
[82] Not much is known about the nitrifier denitrification pathway, but it is believed to be similar to denitrification (Oenema *et al.* 2005).

energy crop demand[83]) will be, in terms of warming potential[84], more important than C losses from land conversion. Similarly, Klemedtsson & Smith (2011) showed that for Northern Europe, there is more than 50% chances that biofuels production will contribute to exceed 54.5 g CO_2 eq. MJ^{-1}, because of N_2O emissions (54.5 g CO_2 eq. MJ^{-1} corresponds to 35% GHG reduction in comparison to the fossil petrol/diesel default reference, based on the Renewable Energy Directive[85]).

In the perspective of a high bioenergy future, several questions thus remain to be answered regarding the potential importance of N_2O. What will be the portion of induced cultivation occurring on organic soils, where N_2O emissions much greater than those estimated with the IPCC methodology (IPCC 2006c) are likely? How to best estimate the N_2O emissions occurring in different biomes of the world, where site-specific relationships like those described in Thomsen *et al.* (2010) may be difficult to represent? What is the most appropriate uncertainty range to use, and which estimation methodologies should be preferred (top-down like Crutzen *et al.* 2007 or bottom-up like IPCC 2006c)?

10.4.4 ILUC

Table 13 and Table 14 showed a wide range of values with respect to the global warming impact of indirect land use changes. This highlights the considerable uncertainty related to the estimation of ILUC, which has been emphasized in several publications already (Plevin *et al.* 2010; Khanna & Crago 2012; Sanchez *et al.* 2012). An overview of the numerous uncertainties involved when estimating the overall ILUC effects are summarized in Plevin *et al.* (2010).

It must be highlighted that although its actual magnitude is uncertain, the potentiality of adverse effects arising from indirect land use changes is hardly subject to dispute (Marelli *et al.* 2011; Gawel & Ludwig 2011; Khanna & Crago 2012); in other words, it is well acknowledged that the GHG related to ILUC are not zero, nor insignificant. Disagreements emerge, however, with respect to the quantification of these ILUC effects and to the way these should be incorporated into policies.

In this PhD work, it was demonstrated that the effects of ILUC can cancel out any benefits of bioenergy based on land-dependent biomass. In Tonini *et al.* (***II***), an attempt to reflect the magnitude of the range of ILUC effects on GHG was made, and it was concluded that this range was wide enough to change the conclusions of the LCA (Figure S19, SI of Tonini *et al.* ***II***).

As emphasized in section 4, only two out of the five components of the overall “ILUC effect” were taken into account in this PhD work, namely land expansion and cultivation of the reacting crops after the final land expansion[86]. Intensification, foregone sequestration capacity and the DLUC effects occurring along the displacement-replacement chain (i.e. “displacement effect”) were not accounted for. Accounting for these would, most likely, increase the GHG figures derived for ILUC in Hamelin/Tonini *et al.* (***II, V***), rather than decreasing them.

The approach used in this PhD work to estimate the land expansion occurring as a result of displacing 1 ha of marginal crop in Denmark was indirect, in the sense that it was based upon the output results derived from a general equilibrium model simulating another situation. This situation, i.e. the increase of wheat demand from Denmark, was however considered as an acceptable proxy to model the ILUC response involved in the case studies carried out within this PhD work. Further, it presented the advantage, through the DM yield, to be easily related to the point of departure for ILUC in this PhD work, i.e. the 1 ha of marginal crop displaced

[83] Here, the model considered a “global cellulosic biofuel program”. It is considered that in 2100, 323-365 EJ y^{-1} of cellulosic biofuels would be produced, worldwide.
[84] In their calculations of N_2O emissions, Melillo *et al.* (2009) used the top-down approach of Crutzen *et al.* (2008).
[85] This 35% reduction is, in the Renewable Energy Directive, a threshold limit up to January 1st, 2017, after which it becomes 50%. The default “fossil fuel comparator” value presented by the Directive is 83.9 g CO^2 eq. MJ^{-1}.
[86] Although DLUC was accounted for, it cannot, by definition, be considered as part of the overall ”ILUC effect”. On the other hand, DLUC is part of the overall “LUC effect”.

from Denmark. In future work, however, this rough approach should be refined though directly modeling, with a general or partial equilibrium model, the ILUC effect of displacing 1 ha of marginal crop in Denmark. This exercise should then be performed for a variety of marginal crops. Based on this, as well as on country-specific crop inventories, the displacement effect could then be modeled. It can however be argued that such econometric models present the drawback to be little transparent. It is in fact very difficult, with these models, to report the exact system boundary used and thus to point out clearly what effects are included and what effects are not[87]. On the other hand, econometric models seem to be the best available alternative allowing to capture the complex interactions resulting from changes in land use, including the price elasticity effects, and the overall induced decreases in crop consumption, among others. Insights allowing to improve these models opacity, user-friendliness, as well as their overall ability to model ILUC[88] would nevertheless be a great benefit to the LCA community.

As mentioned in section 6.4, the flows of C resulting from soil CH_4 uptake have not been included for the DLUC database. This was also not considered for land use changes, although the change in CH_4 uptake capacity when an unmanaged soil is converted to an agricultural soil may in fact involve an overall increase of CH_4 in the atmosphere. On the other hand, none of the published ILUC studies to date (to author's awareness) have included this effect. Nevertheless, including this effect would contribute to increase the overall ILUC GHG figure.

For intensification, Edwards *et al.* (2010) suggested, based on an analysis of the crop and fertilizer prices for UK wheat and US maize, that the "marginal" N use per tonne of crop may be roughly 5 to 12 times higher than the "average" N use per tonne of crop. Of course, using this interval (i.e. application doses 5 to 12 times higher than "average" doses) for estimating the GHG related to intensification would likely result in an overestimation, as the intensification response will, in practice, not be due to an increase of fertilizers input only (section 4.3.3). Further, this approach requires a precise knowledge of the intensification response in terms of i) where it happens and ii) which crops are concerned, although the work of Kløverpris (2008), upon which the estimation of the ILUC response was based in this PhD work, provides these pre-requisite parameters. Another approach that could have been used is the one proposed by Stehfest *et al.* (2010), where an average emission factor of 0.2 to 0.4 t CO_2 eq. ha^{-1} intensified was derived based on a linear relationship between the observed yields and N application rates from 1970 to 2000, worldwide. Again, this however requires that the share of the intensification response, in terms of ha intensified, is known. At the light of the results obtained in Hamelin *et al.* (***I***), the emission factors proposed by Stehfest *et al.* (2010) however appear little realistic. In fact, the overall N_2O for e.g. "average" Danish spring barley cultivation was estimated to ca. 1.2 t CO_2 eq. ha^{-1}, which is already three times greater than the emission factors of Stehfest *et al.* (2010). Yet, the N_2O emissions resulting from intensification are expected to be globally higher than those of "average cultivation". Nevertheless, in the light of the simulation carried out by Melillo *et al.* (2009), where fertilization-related N_2O contributed more to the ILUC effect than the CO_2 emissions resulting from land expansion, it appears obvious that an attempt to quantify the GHG impact of intensification should be made in future work. To this end, the above-described approach of Edwards *et al.* (2010) could be used, where a range of marginal N use per tonne of crop could be derived for a variety of affected crops.

As mentioned in section 4.3.2, the environmental impacts of displacement have typically been disregarded in the ILUC studies published so far. Likewise, it has not been taken into account in this PhD work. The main challenge for doing so is the availability of quality datasets (particularly soil C changes), such as those presented in Hamelin *et al.* (***I***), for all crop and biome systems where displacement-replacement occurs.

[87] Personal communication with David Laborde, February 2013.
[88] Weaknesses that could be improved are for example highlighted in Marelli *et al.* (2011).

10.4.5 Marginal energy

Until recently, a 100 % renewable Danish energy system was still something seen as a distant future, and in the LCA community, the use of marginal electricity did not consider the issues of fluctuating power production, nor the implication that different timeslots of electricity supply and demand can have different marginals[89]. Likewise, this was not applied in the LCA cases included in this PhD thesis. In the context of the recent common energy agreement of the Danish Parliament (Danish Government 2012), it has become clear that a future with a high degree of fluctuating wind power (which implies differences in underlying power sources in periods of high wind versus low wind), is not that distant anymore. Already in 7 years time (in 2020), Denmark will, on the basis of the above-mentioned agreement, have at least 50% of its electricity consumption as wind power. This acknowledgement however came too late to influence the choice of marginal electricity used in the case studies modeled within this PhD work.

However, future LCA models should i) quantify the mix “wind-biomass” energy marginal involved in a future Danish renewable energy systems including ca. 50% wind power and ii) determine the biomass marginal as well as its environmental impact.

10.4.6 Future framework conditions

Future framework conditions surely represent a tremendous uncertainty to be addressed. For example, how would the ILUC effect be affected by a dramatic increase of bioenergy worldwide? Once all the most competitive lands are under cultivation, what would be the environmental cost of turning one more hectare of land to agriculture, or to intensify one more hectare of land? And what will be the result of the ILUC impact in terms of biodiversity loss? Further, phosphorus availability, a scarce and non-substitutable resource (Cordell *et al.* 2009; Neset & Cordell 2012; Seyhan *et al.* 2012), may be limited in the future. How would this affect bioenergy production and crops yields worldwide? Similarly, will the well-documented pollinator decline continue (e.g. Klein *et al.* 2007; Potts *et al.* 2010; Carvalheiro *et al.* 2010), and if so, how will this affect worldwide yields? How will these changing framework conditions interact with the amount and type of food/feed demanded worldwide, or with the changing climate?

These are a few examples of future framework conditions that could potentially strengthen the conclusion that bioenergy based upon land-dependant biomass should be minimized as much as possible. This highlights the relevance for future research on technologies/strategies allowing to reduce the need for such biomass in future renewable energy systems, in Denmark and beyond.

[89] In the light of sections 10.1 and 2.4.1, wind is in fact likely to be the marginal electricity in the periods of excess wind production, while this would rather be biomass in the periods where the electricity demand is greater than the wind power produced.

11. CONCLUSION

In the light of the specific objectives elaborated in section 1, it can be concluded that the objectives of this PhD work have been reached. In fact:

1) A Danish-specific life cycle inventory database comprising 528 different combinations of annual and perennial crops, climates, soil types, initial soil C level, residues management and soil C turnover rate was established. For all these combinations, the input and output flows from and to the environment were quantified, including soil carbon changes. Further, the established database documents the partition of the DM, C and N flows between the primary yield, secondary yield, above- and below-ground residues for all combinations addressed. This consequential LCA database is rather innovative for including such a high level of details, for including soil C changes and for including crops like willow and *Miscanthus* for which, to author' knowledge, no LCI database are yet available. Moreover, it is a valuable and essential input for assessing the environmental consequences of different bioenergy scenarios and conversion routes to be involved in a Danish fossil-free energy system.

2) In the light of the most promising energy crops highlighted in the above-mentioned database, a bioenergy case study addressing 12 different combinations of perennial crops (*Miscanthus*, willow and ryegrass) and conversion technologies (anaerobic co-digestion with manure, thermal gasification, combustion in small-to-medium scale CHP plants and co-firing in large scale coal-fired CHP plants) was performed. This LCA case study used the database mentioned in 1) to address the direct land use change consequences of these bioenergy scenarios, and further included indirect land use change consequences. This case study represents a significant step forward, in comparison to existing bioenergy LCAs, in the sense that it includes both direct and indirect land use changes, and transparently documents the fate of all C and N flows involved in the system, from cultivation to energy production. In this sense, it represents a methodological platform that can be used, and further developed, by LCA practitioners for improving the quality of bioenergy LCAs.

3) A methodology allowing to handle the main challenges posed by manure systems, i.e. the dependency of emissions, at any point of the system, upon the manure composition, was developed. The essence of the developed methodology consisted of a step-wise procedure to define a reference manure management system, including the establishment of a reference manure composition ex-animal, ex-housing and ex-storage that is consistent with the input and output substance flows to and from the manure continuum. Mineral fertilizers substitution was also tackled. This methodology proved to be essential for performing LCAs involving manure (point 4).

4) On the basis of the methodology developed in 3), two LCA case studies assessing the environmental consequences of different strategies for supplying a drastic increase of manure-biogas in Denmark were performed. The first case study investigated the possibility of increasing manure-biogas without relying on the availability of external carbon co-substrates. It consisted to co-digest raw pig and cattle slurry together with the concentrated solid fraction resulting from (ex-housing) manure separation. Three scenarios were assessed, each considering different slurry separation technologies to obtain the solid fraction input for biogas production. In the second case study, additional options were investigated, with a focus on external C co-substrates. Five external co-substrates not already fully used for biogas were considered: energy crops (represented by maize silage), straw, household biowaste, commercial biowaste and garden waste. Further, the use of the solid fraction deriving from source-segregation of animal urine and feces was also investigated. This latter option differed from the first case study, as it involved a separation system directly under the animals, where the contact between feces and urine is prevented at the first place. These six

scenarios were also compared with a mono-digestion case. In the light of Denmark's target to digest 50% of its manure by 2020, these case studies provided timely answers.

The main results of this PhD work can be summarized as follows:

- The market-driven land expansion (i.e. indirect land use change) resulting from using more Danish arable land for energy crop cultivation was shown to offset any potential benefits of bioenergy, unless high-yielding crops (i.e. >10 t DM ha^{-1}) with low water content or low DM losses were considered, in combination with efficient conversion technologies (i.e. 85-90%). The indirect land use changes impact was quantified as 310±170 and 357±195 t CO_2 ha^{-1} displaced, depending on the yield considered for the crop displaced by energy crop in Denmark. Other components of indirect land use changes, like the impacts of intensification, or the foregone sequestration capacity of natural vegetation, were not included.

- All anaerobic co-digestion scenarios highlighted the important environmental benefits, particularly with respect to global warming, of avoiding the reference manure management to take place, i.e. its conventional storage and land application without further processing. As a result, important additional benefits were obtained for the scenarios allowing to use more manure for co-digestion. This finding also emphasized that manure, in a Danish renewable energy system, should be prioritized for biogas.

- The environmental benefits of using separated solid manure (ex-housing) as a co-substrate to boost the methane production of raw slurry were highly dependent upon the efficiency of the separation technology used to concentrate the volatile solids in the solid fraction. Yet, this biogas production concept appeared as limited in the perspective of a wide-spread strategy for increased manure-biogas. In fact, acknowledging that global warming is a key concern, only one of the studied alternatives allowed for clear GHG reductions compared to the reference slurry management. Yet, this alternative involved the use of a polymer, namely cationic polyacrylamide (PAM), which likely persists and accumulates in the environment, and which does represent a potential toxicity risk, although this could not be quantified in the LCA. On this basis, further research on efficient separation technologies not involving cationic PAM appears necessary.

- Source-segregated solid manure (i.e. obtained from preventing any contact between urine and feces) was highlighted as the co-substrate yielding the greatest environmental benefits overall. This mostly reflected that it allowed to use a lot more manure for biogas than the other scenarios. Although this scenario appeared to be favorable for the long-term, it may not be so realistic to rely on this carbon co-substrate in the short-term perspective (2020), as it would involve major changes and investments in current farm buildings.

- Straw and biowastes (i.e. garden waste as well as household and commercial food waste) should be prioritized for manure-biogas, rather than for their other potential uses (i.e. thermal energy recovery and composting). The rationale for this is that the use of these co-substrates for biogas:
    - Resulted in a lower global warming potential than their use for incineration and composting;
    - Allowed to recycle these co-substrate's nutrients, including the slowly degradable carbon, which are essentially lost in the incineration case;
    - Produces a storable gas that can be used for both CHP, transport, and the synthesis of C-based material/chemicals, a key flexibility asset for a renewable energy system involving more than 50% wind power;

    - Allows to avoid energy crops to be used for manure-biogas, and thereby the indirect land use changes related to it.

- Energy crops, because of the land use change they generate, should be used as a last priority within a renewable energy system. However, to the extent they are needed, long duration perennial crops (i.e. *Miscanthus* and willow) should be favored. Particularly, *Miscanthus* was highlighted as the most promising of the investigated energy crops, as it has a relatively high yield, the lowest emission flows of nitrogen compounds, involves relatively low losses of N and P towards aquatic recipients, and allows to increase soil organic carbon. Results however showed that the magnitude of these benefits depends on the harvest season, soil types and climatic conditions.

- Winter wheat was highlighted as the only annual crop where straw removal for bioenergy may be suitable, being the only annual crop not involving losses of soil organic carbon as a result of harvesting the straw. This, however, was only true for sandy soils, and was conditional to manure application. On this basis, and in the light of on-going work on assessing the quality of Danish soils, straw removal should preferably take place on soils with low clay-to-SOM ratio (i.e. <10). Such soils cover most of Jutland, but are also found on Funen and Zealand.

- Finally, it was pointed out that, in a renewable energy future, biomass will become the main source of carbon. In this respect, it was emphasized that carbon efficiency of future biomass & technology combinations will be a decisive concern in a fossil free society.

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Paper I

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Modelling the carbon and nitrogen balances of direct land use changes from energy crops in Denmark: a consequential life cycle inventory

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Abstract

This paper addresses the conversion of Danish agricultural land from food/feed crops to energy crops. To this end, a life cycle inventory, which relates the input and output flows from and to the environment of 528 different crop systems, is built and described. This includes seven crops (annuals and perennials), two soil types (sandy loam and sand), two climate types (wet and dry), three initial soil carbon level (high, average, low), two time horizons for soil carbon changes (20 and 100 years), two residues management practices (removal and incorporation into soil) as well as three soil carbon turnover rate reductions in response to the absence of tillage for some perennial crops (0%, 25%, 50%). For all crop systems, nutrient balances, balances between above- and below-ground residues, soil carbon changes, biogenic carbon dioxide flows, emissions of nitrogen compounds and losses of macro- and micronutrients are presented. The inventory results highlight *Miscanthus* as a promising energy crop, indicating it presents the lowest emissions of nitrogen compounds, the highest amount of carbon dioxide sequestrated from the atmosphere, a relatively high carbon turnover efficiency and allows to increase soil organic carbon. Results also show that the magnitude of these benefits depends on the harvest season, soil types and climatic conditions. Inventory results further highlight winter wheat as the only annual crop where straw removal for bioenergy may be sustainable, being the only annual crop not involving losses of soil organic carbon as a result of harvesting the straw. This, however, is conditional to manure application, and is only true on sandy soils.

Keywords: bioenergy, carbon, direct land use changes, life cycle inventory, nitrogen, straw

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Introduction

The Danish Government has set a long term strategy for Denmark to be independent of fossil fuels in 2050, and several studies have been conducted to design and optimize such a system (Danish Commission on Climate Change Policy, 2010; Energinet.dk, 2010; Lund *et al.*, 2011). These studies all point to the need for a biomass potential of around 35–50% of the overall energy consumption, being 300–450 PJ y^{-1} of biomass out of Denmark's present 850 PJ y^{-1} overall energy consumption. Yet, Denmark is a country with a rather high availability of biomass residues, given it is one of the world's most intensively farmed countries. Despite of that, a biomass supply of 300–450 PJ y^{-1} cannot be met by the circa 200 PJ of biomass residues from agriculture, forestry, industry and households generated in Denmark every year. To provide the necessary biomass feedstock for a Danish fossil free society, conversion of agricultural land from food/feed crops to energy crops, would, therefore, be necessary, if no significant import dependency of biomass is accepted.

This study addresses the environmental consequences of such conversion of agricultural land from food/feed crops to energy crops. These consequences fall into two categories, often named *direct land use changes* (dLUC) and *indirect land use changes* (iLUC). While iLUC refers to the market forces-driven land use changes occurring as a reaction to food/feed displacement on the food/feed market (Edwards *et al.*, 2010; Searchinger, 2010), dLUC represents the change in the land use allocation of a given country or region that caused this displacement to occur in the first place (e.g. allocating more Danish land nowadays used to grow food/feed crops to energy crops). This article addresses the dLUC only.

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The objective of this study is to develop a consequential life cycle inventory (LCI) for assessing the dLUC consequences of converting Danish agricultural land from food/feed crops to energy crops. Though LCI databases already exist for some crops (e.g. Ecoinvent), these apply for specific countries that may not fully represent the Danish situation, given that many of the emission flows involved are rather site-specific, and that the management of the crop systems (e.g. fertilizer input) differs, among others because of the differences in legislation. Moreover, none of the existing LCI databases address all the following issues, which are addressed in the present study:

- The partition of biomass between above- and below-ground biomass, and consequently the partition of the C and N contained in the biomass between these different fractions. This is very seldom taken into account into life cycle assessments (LCAs), and often completely ignored (Larson, 2006), although the need of it is increasingly recognized and claimed (e.g. Cherubini *et al.*, 2009; Börjesson & Tufvesson, 2011; Brandão *et al.*, 2011), especially in the light of including soil C changes into LCA. This is also intrinsically necessary in order to determine an appropriate balance between the residues that should be returned to the soil for maintaining soil fertility, and the amount that can be removed for energy production. Scientific basis to determine such a balance has actually been identified as a key research need for establishing sustainable bioenergy systems (Bringezu *et al.*, 2009).
- Soil C changes. In most LCAs, the C balance is incomplete, the C uptake from the atmosphere being assumed equal to the C harvested plus the C released from decay of plant residues. Yet, these flows are not necessary equal, and a correct balance should take into account the amount of C sequestrated/released from the soil, over the time horizon considered.
- Perennial crops. Though comprehensive LCA inventories do exist for some annual crops (e.g. Jungbluth *et al.*, 2007; Nemecek & Kägi, 2007), very few, if any, complete LCA datasets are available for perennial crops like *Miscanthus* and willow, albeit LCAs on *Miscanthus* and willow do exist as well as datasets for some grass types.

Material and methods

Overview of the inventory structure

As a first step of this LCI, the most influential parameters on the biogeochemical flows of C and N for which a specific inventory was judged necessary were identified. As a result, a considerable level of details has been included in the inventory, resulting in a total of 528 combinations, for which the input and output flows from and to the environment are quantified, including soil C changes. The variables and sub-variables considered are illustrated in Fig. 1. As shown in Fig. 1, the inventory includes four annual and three perennial crops and distinguishes between spring and autumn harvest of *Miscanthus*. The reason for this is that the harvest season involves different trade-offs and is likely to influence the conversion route. For example, while the dry matter (DM) yield is higher if harvest occurs in autumn, delaying the harvest to spring involves a lower concentration of minerals in the harvested biomass which favours a better combustion quality (Jørgensen, 1997; Lewandowski & Heinz, 2003), besides supplying a higher input of C to the soil due to leaves and tops losses occurring during winter. Similarly to *Miscanthus*, spring barley is treated in two different ways (Fig. 1) in order to assess the environmental consequences of combining it with a catch crop, which is used for 0.12–0.20 Mha of the Danish agricultural area every year. This consists of a non-commercial crop (e.g. perennial ryegrass, oilseed radish) grown to catch the available N in the soil during the autumn period, thereby reducing N leaching (Thorup-Kristensen *et al.*, 2003). Though it appears twice in Fig. 1, there is only one scenario for willow. This is because willow, as opposed to the other crops, can only be fertilized with either 100% slurry (harvest years) or 100% mineral fertilizers (other years), since slurry spreading is only possible in harvest years with the currently available equipment, under Danish conditions.

Perennial crops life cycle

As opposed to annual crops that are simply sown and harvested every year, perennial crops have a more complex life cycle, involving different activities for the different growth years. The life time considered for a *Miscanthus* plantation in this study is 20 years (18 cuts; 1 year establishment: 1 year preparation before planting). For willow, a 21 years life cycle is considered (six cuts; 3 years harvest cycle; 1 year establishment; 1 year preparation before planting). The different activities considered throughout each year of both *Miscanthus* and willow life cycles are further detailed in the Supporting Information (Appendix S1). The life cycle considered for perennial ryegrass is 2 years, which is common practice in Danish agriculture; sowing here occurs every second year, but harvests take place annually.

Agricultural operations

Besides fertilization, eight main categories of agricultural operations are considered, namely soil preparation, propagation (seed, rhizome or cutting production), liming, sowing/planting, plant protection, irrigation, harvest and transport from farm to field. For each of these operations, the modelling details (e.g. specific processes used, quantities considered or diesel consumption) are presented in the Supporting Information (Appendix S2).

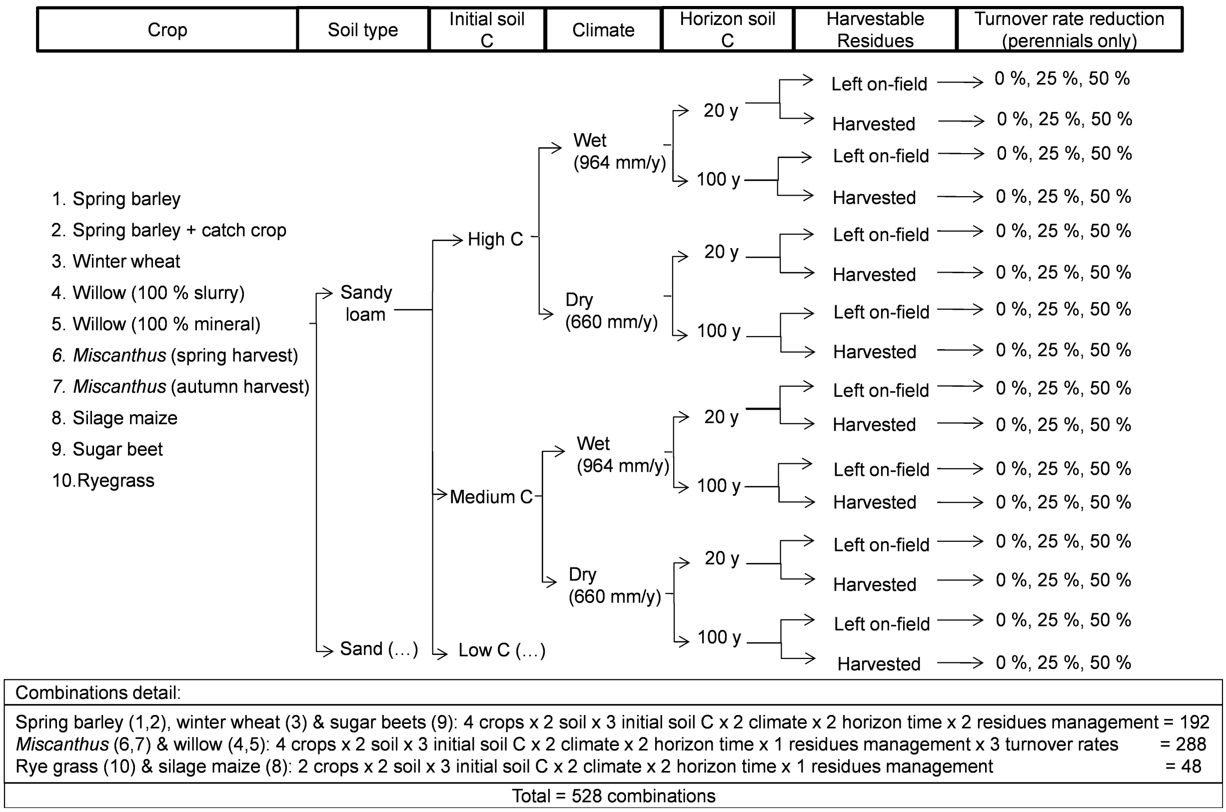

Fig. 1 Overview of the variables selected for the inventory structure.

Fertilization

The fertilization operations are performed in conformity with Danish regulations (Danish Ministry of the Environment, 2006; Danish Ministry of Food, Agriculture & Fisheries, 2008), involving an upper ceiling for the amount of N to be applied on the field. Based on the statistics presented by Nielsen *et al.* (2009) on the N applied to Danish soils from 1990 to 2007, it is assumed that the N demand of the crops is fulfilled by 50% mineral fertilizers and 50% animal manure (further divided into 50% fattening pig slurry, and 50% dairy cattle slurry, as described in Appendix S2). This does not apply for willow, which is whether fertilized by 100% mineral fertilizers or by 100% animal manure, as earlier described. The mineral N, phosphorus (P) and potassium (K) fertilizers considered are calcium ammonium nitrate, diammonium phosphate and potassium chloride, respectively. The annual fertilization needs for each of the selected crops in terms of N, P and K, which vary according to soil types, are presented in the Supporting Information (Appendix S2). For all crops, most of the P and K needs are fulfilled by the added slurry. However, when the balance is not fulfilled, it is assumed that mineral fertilizers are applied to fully fulfil crop requirements. The fertilization balance for each crop and soil type is presented in Table 1.

DM partitioning between harvest yield and residues

In this study, the DM from crop products is separated into four main categories: primary yield, secondary yield, non-harvestable DM above-ground (e.g. stubble, leaves, branches and twigs from woody crops, etc.), and non harvestable DM below-ground (root residues).

Primary crop yield refers to the yield of the main product, i.e. the product motivating the cultivation. For annual crops and ryegrass, the primary yields used in this study are those presented in the national fertilization guidelines (Danish Ministry of Food, Agriculture & Fisheries, 2009), so that consistency is ensured with the fertilization input considered. Primary yield data for willow were adapted from Lærke *et al.* (2010) and from Olesen *et al.* (2001) for spring and autumn harvested *Miscanthus* (Appendix S3).

Secondary yields consist of the harvestable fraction of crop residues and consequently do not include fractions that are non-harvestable (e.g. part of the straw that cannot be harvested due to machinery-related constraints). In this study, secondary yields apply for spring barley (with and without catch crop), winter wheat and sugar beet, and were estimated based on the ratio between harvested residues and harvested crops from Statistics Denmark (2010). Based on this, the ratio between secondary yield and primary yield was considered to be 0.55 for spring barley and winter wheat and 0.13 for sugar beet.

Knowing the primary as well as the secondary yields, the non-harvestable DM from above-ground residues can be estimated from the harvest index (HI), for the crops for which the HI was available. The HI represents the primary yield share of total above-ground biomass (primary yield, secondary yield and non-harvestable residues) at harvest. Harvest indexes

Table 1 Annual fertilizer input, for all crops, on sandy and sandy loam soils*. All values are expressed per ha per y, and apply for both dry and wet climates

Crops	Spring barley		Spring barley & catch crop		Winter wheat		Willow (100% slurry)†	Miscanthus (autumn harvest)	Miscanthus (spring harvest)	Maize silage		Sugar beet		Ryegrass		Willow (100% mineral)†	Miscanthus (both), Year 2	Miscanthus (both), Year 3
Soil type	Sand	Sandy loam	Sand	Sandy loam	Sand	Sandy loam	Sand and sandy loam	Sand and sandy loam	Sand and sandy loam	Sand	Sandy loam	Sand	Sandy loam	Sand	Sandy loam	Sand and sandy loam	Sand and sandy loam	Sand and sandy loam
Pig slurry‡																		
Amount (ton)	8.8	8.0	7.6	6.8	11.6	11.3	16.8	6.3	4.2	11.3	9.7	8.6	7.1	23.9	22.7	0	2.1	4.2
N (kg)	42.0	38.0	36.3	32.3	55.3	53.7	80.0	30.0	20.0	54.0	46.3	41.0	34.0	114.0	108.3		10.0	20.0
P (kg)	8.8	8.0	7.6	6.8	11.6	11.3	16.8	6.3	4.2	11.3	9.7	8.6	7.1	24.0	22.8		2.1	4.2
K (kg)	23.1	20.9	20.0	17.8	30.5	29.5	44.0	16.5	11.0	29.7	25.5	22.6	18.7	62.7	59.6		5.5	11.0
C (kg)	257.5	233.0	222.8	198.3	339.3	329.1	490.6	184.0	122.6	331.1	284.1	251.4	208.5	699.1	664.3		61.3	122.6
Cu (kg)	0.2	0.2	0.2	0.2	0.3	0.3	0.5	0.2	0.1	0.3	0.3	0.2	0.2	0.7	0.6		0.1	0.1
Zn (kg)	0.7	0.7	0.6	0.6	1.0	0.9	1.4	0.5	0.3	0.9	0.8	0.7	0.6	2.0	1.9		0.2	0.3
Cattle slurry§																		
Amount (ton)	7.8	7.0	6.7	6.0	10.2	9.9	14.8	5.5	3.7	10.0	8.6	7.6	6.3	21.0	20.0	0	1.9	3.7
N (kg)	45.0	40.7	38.9	34.6	59.3	57.5	85.7	32.1	21.4	57.9	49.6	43.9	36.4	122.1	116.1		10.7	21.4
P (kg)	7.7	6.9	6.6	5.9	10.1	9.8	14.6	5.5	3.7	9.9	8.5	7.5	6.2	20.8	19.8		1.8	3.7
K (kg)	45.2	40.9	39.1	34.8	59.5	57.7	86.1	32.3	21.5	58.1	49.9	44.1	36.6	122.7	116.6		10.8	21.5
C (kg)	350.4	317.0	303.1	269.8	461.6	447.7	667.4	250.3	166.9	450.5	386.6	342.1	283.7	951.1	903.8		83.4	166.9
Cu (kg)	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.1	0.04	0.1	0.1	0.10	0.1	0.2	0.2		0.02	0.04
Zn (kg)	0.2	0.2	0.1	0.1	0.2	0.2	0.3	0.1	0.1	0.2	0.2	0.2	0.1	0.5	0.4		0.04	0.1
Mineral N																		
N (kg)	63.0	57.0	54.5	48.5	83	80.5	0	45.0	30.0	81.0	69.5	61.5	51.0	171.0	162.5	120.0	15.0	30.0
Mineral P																		
P (kg)	5.5	7.1	7.7	9.3	0.3	0.9	0	3.2	7.2	22.8	25.8	26.9	29.7	0	0	15.0	11.1	7.2
Mineral K																		
K (kg)	0	0	0	0	0	0	0	26.2	42.5	47.2	59.7	83.3	94.7	53.6	62.8	50.0	58.7	42.3
Total nutrients added																		
N (kg)	150.0	135.7	129.7	115.5	197.6	191.7	165.7	107.1	71.4	192.9	165.5	146.4	121.4	407.1	386.9	120.0	35.7	71.4
P (kg)	22.0	22.0	22.0	22.0	22.0	22.0	31.4	15.0	15.0	44.0	44.0	43.0	43.0	44.8	42.5	15.0	15.0	15.0
K (kg)	68.3	61.8	59.1	52.6	90.0	87.3	130.1	75.0	75.0	135.0	135.0	150.0	150.0	239.0	239.0	50.0	75.0	75.0
C (kg)	607.9	550.0	525.9	468.0	800.9	776.8	1158.0	434.2	289.5	781.6	670.7	593.5	492.1	1650	1568	0.0	144.8	289.5

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Table 1 (continued)

Crops	Spring barley		Spring barley & catch crop		Winter wheat		Willow (100% slurry)†	Miscanthus (autumn harvest)	Miscanthus (spring harvest)	Maize silage		Sugar beet		Ryegrass		Willow (100% mineral)†	Miscanthus (both), Year 2	Miscanthus (both), Year 3
Soil type	Sand	Sandy loam	Sand	Sandy loam	Sand	Sandy loam	Sand and sandy loam	Sand and sandy loam	Sand and sandy loam	Sand	Sandy loam	Sand	Sandy loam	Sand	Sandy loam	Sand and sandy loam	Sand and sandy loam	Sand and sandy loam
Cu (kg)	0.3	0.3	0.3	0.3	0.4	0.4	0.6	0.2	0.2	0.4	0.4	0.3	0.3	0.9	0.9	0.0	0.1	0.2
Zn (kg)	0.9	0.8	0.8	0.7	1.2	1.1	1.7	0.6	0.4	1.2	1.0	0.9	0.7	2.4	2.3	0.0	0.2	0.4

*This table presents the annual fertilization needs for all crops, though for *Miscanthus*, there is no fertilization on year 1 of the life cycle, and for willow, on years 1 and 2.

†There is only one willow scenario, not 2. However, since the fertilization balance is different for the years with 100% animal slurry fertilizers vs. the years with 100% mineral fertilizers, both are shown in the table.

‡Considering an efficiency of 75% for fattening pig slurry N, i.e. if 1 kg of slurry-N is applied, only 0.75 kg will be available for plants uptake.

§Considering an efficiency of 70% for dairy cow slurry N, i.e. if 1 kg of slurry-N is applied, only 0.70 kg will be available for plants uptake.

applied in this study are 0.45 for spring barley, 0.45 for winter wheat, 0.70 for sugar beet and 0.85 for silage maize, based on Gyldenkærne *et al.* (2007) as well as on Olesen *et al.* (2000). For perennial ryegrass, an above-ground residues input of 4.15 Mg DM ha^{-1} y^{-1} is calculated, based on an overall C input to soil estimated to 5.6 t C ha^{-1} y^{-1} (Petersen, 2010), and on the assumption that below-ground and above-ground residues are distributed according to a ratio 2 : 1 (IPCC, 2006). Above-ground residues for *Miscanthus* were estimated from the data presented by Olesen *et al.* (2001), and willow's above-ground residues were estimated based on the model developed by Lindroth & Båth (1999). All estimates made for above-ground residues of perennial crops are further detailed in the Supporting Information (Appendix S4).

Below-ground residues produced for annual crops systems were estimated through the use of the ratio of below-ground biomass to total net biomass production (i.e. primary yield, secondary yield and all non-harvestable residues). This ratio was taken from Gyldenkærne *et al.* (2007) and is 0.17 for spring barley, 0.25 for winter wheat, 0.12 for sugar beet and 0.15 for silage maize. For ryegrass, it was considered that there are two times more below-ground residues than above-ground residues (IPCC, 2006). For *Miscanthus*, the annual below-ground biomass was estimated as 16% of the total above-ground biomass (residues plus primary yield; Olesen *et al.*, 2001). The procedure used to estimate below-ground residues of willow is partly based on the model developed by Lindroth & Båth (1999), as detailed in the Supporting Information (Appendix S4).

For catch crop DM input to soil, an amount of 1.40 Mg DM ha^{-1} is assumed, based on the results from Petersen *et al.* (2011) and Chirinda *et al.* (2012). This includes the whole DM input from the catch crop (i.e. the catch crop itself as well as all above- and below-ground residues). This estimate is not meant to represent a specific catch crop, but only to reflect the inclusion of a catch crop on the overall C and N balance in a life cycle perspective.

The DM partition considered for all selected crops, soil types and climates is presented in Table 2.

C and N returned to soil from non-harvested residues

The interest of determining the DM partition from the biomass (primary yield, secondary yield, above-and below-ground residues) lies in estimating the flows of C and N of these different fractions to the environment, for each of the selected crop systems. For C, this is straightforward, as it is assumed, based on Petersen (2010), that the C content of the DM corresponds to 0.45 t C t^{-1} DM for all crop parts. In the case of below-ground residues, this also includes C input from rhizodeposition.

For N in above-ground residues of ryegrass and annual crops (sugar beet excluded), values from the Danish Inventory report to the United Nations Framework Convention on Climate Change (UNFCCC) were used as the best available data (Nielsen *et al.*, 2009). For sugar beet, a value of 0.026 kg N kg^{-1} DM was used (Møller *et al.*,2000; IPCC, 2006). The N content of below-ground residues was estimated for ryegrass and annuals crops from IPCC (2006), as no better data for the specific Danish conditions were available. The N content of above- and

Table 2 Partition of the DM for all selected crops, soils and climates. All values in Mg DM ha^{-1} y^{-1}

DM repartition (Mg DM ha^{-1} y^{-1})	Soil type	Climate	Spring barley (R)*	Spring barley (I)*	Spring barley & catch crop (R)*	Spring barley & catch crop (I)*	Winter wheat (R)*	Winter wheat (I)*	Willow†	*Miscanthus* (autumn) Year 4–20‡	*Miscanthus* (spring) Year 4–20‡	Maize silage	Sugar beet (R)*	Sugar beet (I)*	Ryegrass
Primary yield	Sand§	Wet	4.25	4.25	4.25	4.25	5.61	5.61	10.60	15.25	10.0	12.05	12.45	12.45	9.98
		Dry	4.25	4.25	4.25	4.25	5.61	5.61	7.10	12.96	8.50	12.05	12.45	12.45	9.98
	Sandy loam	Wet	4.85	4.85	4.85	4.85	6.80	6.80	12.72	15.25	10.0	11.93	12.45	12.45	8.82
		Dry	4.85	4.85	4.85	4.85	6.80	6.80	10.81	15.25	10.0	11.93	12.45	12.45	8.82
Secondary yield	Sand§	Wet/dry	2.34	2.34	2.34	2.34	3.09	3.09	0	0	0	0	1.62	1.62	0
	Sandy loam	Wet/dry	2.67	2.67	2.67	2.67	3.74	3.74	0	0	0	0	1.62	1.62	0
Above-ground residues¶	Sand§	Wet	2.86	5.19	2.86**	5.19**	3.77	6.86	5.03	5.63	10.88	2.13	3.72	5.34	4.15
		Dry	2.86	5.19	2.86**	5.19**	3.77	6.86	3.37	4.79	9.25	2.13	3.72	5.34	4.15
	Sandy loam	Wet	3.26	5.93	3.26**	5.93**	4.57	8.31	6.03	5.63	10.88	2.11	3.72	5.34	4.15
		Dry	3.26	5.93	3.26**	5.93**	4.57	8.31	5.13	5.63	10.88	2.11	3.72	5.34	4.15
DM input from catch crop	Sand§	Wet/dry	0	0	1.4	1.4	0	0	0	0	0	0	0	0	0
	Sandy loam	Wet/dry	0	0	1.4	1.4	0	0	0	0	0	0	0	0	0
Below-ground residues	Sand§	Wet	1.93	1.93	1.93**	1.93**	4.16	4.16	5.21	3.34	3.34	2.50	2.43	2.43	8.30
		Dry	1.93	1.93	1.93**	1.93**	4.16	4.16	3.49	2.84	2.84	2.50	2.43	2.43	8.30
	Sandy loam	Wet	2.21	2.21	2.21**	2.21**	5.04	5.04	6.25	3.34	3.34	2.48	2.43	2.43	8.30
		Dry	2.21	2.21	2.21**	2.21**	5.04	5.04	5.31	3.34	3.34	2.48	2.43	2.43	8.30

*R: straw or top removal; I: straw or top incorporation.

†Values are the same whether the willow plantation is fertilized with 100% slurry or with 100% mineral fertilizers.

‡Values for years 1, 2 and 3 are presented in Appendix S3 (primary yields) and Appendix S4 (above- and below-ground residues).

§Yields on sandy soils consider that the crops are irrigated, except for willow and *Miscanthus*.

¶For annuals, it is calculated as: (primary yield/harvest index) minus primary yield minus secondary yield. For perennials, it is as indicated in the text.

**This does not include the contribution from the catch crop.

below-ground residues for willow was assumed to be 0.005 kg N kg^{-1} DM, based on Eckersten *et al.* (2006). The N content of above-ground residues was assumed to be 0.006 kg N kg^{-1} DM for autumn harvested *Miscanthus* and 0.01 kg N kg^{-1} DM for spring harvested *Miscanthus*, based on Jørgensen (1997). This applies for all life cycle years of *Miscanthus* (i.e. year 1, 2 and 3 and years 4–20). The N content of below-ground residues was taken at 0.005 kg N kg^{-1} DM for both autumn and spring harvested *Miscanthus* based on Neukirchen *et al.* (1999) and Strullu *et al.* (2011), who measured N concentrations in living below-ground biomass of between 0.005 and 0.017 kg N kg^{-1} DM.

For scenarios involving incorporation of secondary yield, the N content of straw and beet tops had to be considered. The N content of spring barley and winter wheat straw was estimated using the content of raw protein in straw from the values presented in Møller *et al.* (2000), i.e. 4.0% of DM for spring barley straw and 3.3% of DM for winter wheat straw. To obtain the N content of straw, it was assumed that the average N content of protein is 16% (FAO, 2003). For sugar beet tops, the N content was taken from Nielsen *et al.* (2009). An overall input of 0.034 kg N kg^{-1} DM was considered for the catch crop.

The balance of C and N returned to soil for all crops, soils and climates considered is presented in Table 3.

N losses

In the cropping systems considered in this study, there are three main input of N: from fertilizers, from crop residues, and from the atmosphere. The output flows considered are ammonia (NH_3), nitrous oxide (N_2O) (direct and indirect), nitrogen oxides (NO_x), emissions of dinitrogen (N_2) to the atmosphere and nitrate (NO_3^-) leaching to ground- and surface waters.

Two NH_3 flows were estimated: the NH_3 from the application of mineral fertilizers and the NH_3 from the application of animal slurry. The emission factors considered for estimating the NH_3 emissions from animal slurry and mineral fertilizer applications are presented in Table 4. Other sources could have been considered in estimating NH_3 flows, namely the decaying crops residues as well as the NH_3 emission from crop foliage. Crop foliage was not included as a source of NH_3 emission due to the contradicting results and evidences found regarding the quantification of this emission and its actual occurrence in arable cropland. Similarly, the NH_3 emission occurring as a result of the decomposition of crop residues was not included, because this emission is practically insignificant when residues are incorporated (De Ruijter *et al.*, 2010), which is the case for the annual crop systems in this study. For perennials, it is considered that when crops shed their leaves, these are already emptied of easily convertible N (primarily at the profit of storage organs) which should also result in negligible NH_3 emissions from the residues.

Nitrogen oxides consist of the sum of nitric oxide (NO) and nitrogen dioxide (NO_2). Once emitted from the soil (mostly as a result of nitrification), NO is quickly oxidized to NO_2 by available oxidants in the atmosphere (typically ozone) (Delon *et al.*, 2008). Though gaseous NO_2 is emitted from biological processes occurring in the soil (Graham *et al.*, 1997), no information has been found on NO_2 emissions from soils in the selected crop systems. Therefore, the emissions of NO are assumed to represent total NO_x. Stehfest & Bouwman (2006) reported NO-N emissions for Europe of 144 Gg y^{-1}, for a N application of 12 812 Gg y^{-1}, for cropland. Based on this, an emission factor of 0.011 kg NO-N per kg N applied can be derived. Similarly, an emission factor of 0.013 kg NO-N per kg N applied was derived for grassland. For crop residues, based on Haenel *et al.* (2010), an emission factor of 0.007 kg NO-N per kg N was used.

The formation of N_2O in crop systems is particularly favoured by partial or transient anaerobic conditions, but also by high concentrations of NO_3^- in the soil solution, by the presence of an available carbon source, and by warm temperatures, among others (Stehfest & Bouwman, 2006; Nieder & Benbi, 2008). Because of this dependence upon such site-specific factors, emissions of N_2O exhibit a very high degree of spatial and temporal variability. Emissions of N_2O from cultivation activities are, for LCI, generally estimated based on extrapolation from field measurements, from biogeochemical models or most commonly calculated based on IPCC guidelines (IPCC, 2006). Based on this methodology, N_2O emissions are assumed to be proportional to the N content of the source material for N_2O emissions. Though critiques have been published about the IPCC methodology (e.g. Jungkunst & Freibauer, 2005; Stehfest & Bouwman, 2006; Smeets *et al.*, 2009), the use of IPCC default factors for estimating N_2O emissions is probably the best methodology to use outside well-characterized areas (Edwards *et al.*, 2008). In Denmark, IPCC default emission factors approximately correspond with measured emissions (e.g. Chirinda *et al.*, 2010a). For these reasons, it is the IPCC methodology (IPCC, 2006) that is applied in this study to estimate the N_2O emissions from the different crop systems. Based on this methodology, an emission factor of 0.01 kg N_2O-N per kg N for synthetic fertilizer, organic fertilizer and crop residue input is considered. A portion of the volatilized N (as NH_3 and NO_x) that is re-deposited will subsequently be emitted as N_2O. Similarly, a portion of the N losses through leaching may be emitted as N_2O. These are referred to as indirect N_2O emissions (IPCC, 2006). The IPCC methodology suggests an emission factor of 0.010 kg N_2O-N per kg NH_3-N plus NO_X-N volatilized and of 0.0075 kg N_2O-N per kg N leaching. The IPCC methodology also suggests to account for N_2O emissions occurring as a result of soil organic matter mineralization, in situations where native soil C is lost due to a change in land use or management. This has not been considered in this inventory, because of the inconsistency in the IPCC (2006) approach that only considers emissions of N_2O from losses of soil organic matter and not reductions in estimated N_2O emissions when soil organic matter is accumulated. In fact, the better soil structure and soil aeration associated with higher soil organic matter levels lead to reduced N_2O emissions (Chirinda *et al.*, 2010b). However, this effect was included as a sensitivity analysis, in order to size the importance of this contribution to the overall N_2O emissions.

Leaching of N is, for ryegrass and annual crops, calculated with the N-LES_4 model (Kristensen *et al.*, 2008), a continuously updated empirical model to predict N leaching from arable land based on more than 1200 leaching studies performed in

Table 3 Input of N and C from residues for all selected crops, soils and climates

DM repartition	Soil type	Climate	Spring barley (R)*	Spring barley (I)*	Spring barley & catch crop (R)*	Spring barley & catch crop (I)*	Winter wheat (R)*	Winter wheat (I)*	Willow†	*Miscanthus* (autumn) Year 4–20	*Miscanthus* (spring) Year 4–20	Maize silage	Sugar beet (R)*	Sugar beet (I)*	Ryegrass
Data (all values in kg N or C per kg DM per year)															
N in above ground residues	Sand	Wet/dry	0.004‡	0.004‡	0.004‡	0.004‡	0.003‡	0.003‡	0.005	0.006	0.010	0.003‡	0.03	0.03	0.006‡
	Sandy loam	Wet/dry	0.003‡	0.003‡	0.003‡	0.003‡	0.002‡	0.002‡	0.005	0.006	0.010	0.003‡	0.03	0.03	0.006‡
N in below ground residues	Sand & Sandy loam	Wet/dry	0.01	0.01	0.01	0.01	0.009	0.009	0.005	0.005	0.005	0.007	0.01	0.01	0.01
N from straw, tops and/or catch crop	Sand & Sandy loam	Wet/dry	0	0.006	0	0.04	0	0.005	0	0	0	0	0	0.04	0
C in all crop parts	Sand & Sandy loam	Wet/dry	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45
Total C and N input from residues (all values in kg N or C per ha per y)															
Total N from residues, above and below ground	Sand	Wet	37.48	52.44	85.08	100.04	54.40	70.69	51.18	50.48§	125.5¶	23.81	130.60	187.30	125.76
		Dry	37.48	52.44	85.08	100.04	54.40	70.69	34.29	42.91§	106.7¶	23.81	130.60	187.30	125.76
	Sandy loam	Wet	41.30	58.38	88.90	105.98	62.33	82.08	61.41	50.48§	125.5¶	23.64	130.60	187.30	125.76
		Dry	41.30	58.38	88.90	105.98	62.33	82.08	52.90	50.48§	125.5¶	23.64	130.60	187.30	125.76
Total C from residues, above and below ground	Sand	Wet	2156	3208	2786	3838	3567	4956	4606	4037**	6399††	2083	2764	3492	5600
		Dry	2156	3208	2786	3838	3567	4956	3086	3431**	5439††	2083	2764	3492	5600
	Sandy loam	Wet	2460	3661	3090	4291	4324	6007	5527	4037**	6399††	2062	2764	3492	5600
		Dry	2460	3661	3090	4291	4324	6007	4698	4037**	6399††	2062	2764	3492	5600

*R: straw or top removal; I: straw or top incorporation.

†Values are the same whether the willow plantation is fertilized with 100% slurry or with 100% mineral fertilizers.

‡Original values from Nielsen *et al.* (2009) are, in kg N ha^{-1} y^{-1}: spring barley: 10.4; winter wheat: 17.0; Silage maize: 6.3; Ryegrass: 26.2.

§Values for year 2 are: 15.53 kg N ha^{-1} y^{-1} (sand, dry climate) and 18.27 kg N ha^{-1} y^{-1} otherwise; Values for year 3 are 26.51 kg N ha^{-1} y^{-1} (sand, dry climate) and 31.19 kg N ha^{-1} y^{-1} otherwise.

¶Values for year 2 are: 35.22 kg N ha^{-1} y^{-1} (sand, dry climate) and 41.43 kg N ha^{-1} y^{-1} otherwise; Values for year 3 are 56.96 kg N ha^{-1} y^{-1} (sand, dry climate) and 67.01 kg N ha^{-1} y^{-1} otherwise.

**Values for year 2 are: 1242 kg C ha^{-1} y^{-1} (sand, dry climate) and 1461 kg C ha^{-1} y^{-1} otherwise; Values for year 3 are 2117 kg C ha^{-1} y^{-1} (sand, dry climate) and 2491 kg C ha^{-1} y^{-1} otherwise.

††Values for year 2 are: 1815 kg C ha^{-1} y^{-1} (sand, dry climate) and 2136 kg C ha^{-1} y^{-1} otherwise; Values for year 3 are 2923 kg C ha^{-1} y^{-1} (sand, dry climate) and 3439 kg C ha^{-1} y^{-1} otherwise.

Table 4 Emission factors considered for estimating ammonia emissions

	Animal slurry (kg NH_3-N kg^{-1} N applied)*				Mineral fertilizers (kg NH_3-N kg^{-1} N applied)†		
Crop type	Pig slurry	Cattle slurry	Pig slurry, loss during application process	Cattle slurry, loss during application process	Urea	Calcium ammonium nitrate	Diammonium phosphate
Ryegrass	0.14	0.19	0.004	0.003	0.21	0.015	0.015
All other crops	0.12	0.16	0.004	0.003	0.12	0.020	0.020

*Emission factors based on Hansen *et al.* (2008).
†Emission factors based on the average of six studies: Harrison & Webb (2001), Mikkelsen Hjorth *et al.* (2006), Nemecek & Kägi (2007), EMEP-EEA (2009), Nielsen *et al.* (2009), Haenel *et al.* (2010).

Denmark during the last 15 years. For *Miscanthus*, N leaching estimates were based on data from Olesen *et al.* (2001), and these estimates are further detailed in the Supporting Information (Appendix S5). Nitrate leaching for willow was estimated to be the same as for *Miscanthus*. For both *Miscanthus* and willow, N leaching has been considered to be highest in the planting year (Mortensen *et al.*, 1998).

C losses

Changes in soil C were estimated with the dynamic soil C model C-TOOL, developed to calculate the soil carbon dynamics in relation to the Danish commitments to UNFCCC. This model is parameterized and validated against long-term field experiments conducted in Denmark, UK and Sweden. Further description of the C-TOOL model is given in Petersen *et al.* (2002) and Petersen (2010). Changes in soil C were estimated over two time horizons: 20 and 100 years. Moreover, an initial 'high', 'medium' and 'low' soil C content were considered (Fig. 1). These levels are based on an average of 143.9 ± 59.2 t C ha^{-1} for sandy soils and 144.7 ± 76.4 t C ha^{-1} for sandy loam soils, for the depth 0–100 cm, the medium level being the average, and the high and low levels corresponding to one standard deviation. For *Miscanthus* and willow, the C turnover rate in the topsoil may be reduced in response to the absence of tillage over many years (Olesen *et al.*, 2001; Chatskikh *et al.*, 2009). In this study, three different turnover rates have been applied for these two crops; no reduction in turnover rate (as for other crops), 25% reduced turnover rate and 50% reduced turnover rate.

The portion of the C input to the soil (i.e. from manure, straw/tops and non-harvestable residues) that does not enter the soil C pool over the time horizon considered is assumed to be lost as a CO_2-C emission to the atmosphere. Similarly, all losses of native soil C are assumed to be transferred to the atmosphere as CO_2-C. The details of these calculations are presented in the Supporting Information (Appendix S6).

Lime input were considered separately; each mole of lime applied to the field has the net potential to contribute to the addition of 1 mole of CO_2 to the atmosphere. This emission was estimated at 0.12 kg CO_2-C kg^{-1} lime applied (IPCC, 2006).

Methane (CH_4) emissions in agricultural fields are typically assumed as negligible (e.g. Hamelin *et al.*, 2011), due to the aerobic conditions found in cultivated mineral soils. Some LCA studies (e.g. Brandão *et al.*, 2011), however, considered a CH_4 emission resulting from the inhibition of atmospheric methane uptake by the soil caused by the cultivation of these soils and the use of N fertilizers. This has not been considered in this study because of the limited information on the various food and bioenergy cropping systems on soil methane oxidation. Also, this effect has been shown to be very minor on the overall greenhouse gas balance of perennial and annual crop systems (Robertson *et al.*, 2000).

Other losses

Biogenic non-methane volatile organic compounds (NMVOC) emitted from photosynthesizing leaves of crops (particularly isoprene and monoterpene) are taken into account in the inventory. The calculation of NMVOC in this study is based on the methodology described in Haenel *et al.* (2010) considering specific emission factors (in kg NMVOC DM^{-1} h^{-1}) for the different crop systems, as detailed in the Supporting Information (Appendix S7).

Phosphorus losses from agricultural soils have been estimated as 5% of the net surplus application, based on Nielsen & Wenzel (2007). For perennial crops, P losses are estimated as 2.5% of the net surplus application, based on Sørensen *et al.* (2010). Copper (Cu) and zinc (Zn) losses, which are of relevance for toxicity-related environmental impacts, were estimated similarly as for P losses, though it was assumed that 100% of the surpluses are lost (Wesnæs *et al.*, 2009). The calculation of P, Cu and Zn losses is detailed in the Supporting Information (Appendix S8).

Though the importance of understanding the fate of K and to some extent of calcium (Ca) on an agronomical perspective is recognized, K and Ca losses towards soils and waters are not flows affecting any of the environmental impacts categories described in the Danish EDIP method for life cycle impact assessment (Wenzel *et al.*, 1997; Hauschild & Potting, 2005; Potting & Hauschild, 2005; Stranddorf *et al.*, 2005). The fate of K and Ca is therefore not considered further in this study.

Sensitivity analyses

The N fertilizer considered in this study is calcium ammonium nitrate (CAN), which applies for a North European scope.

Worldwide, however, urea is by far the most consumed N fertilizer (EFMA, 2009; FAOSTAT, 2010; IFA, 2010), representing 55% of world N fertilizer production capacity in 2006/2007 (EFMA, 2010). Moreover, the use of urea is increasing in Europe (EFMA, 2009). For this reason, and because the differences in environmental impacts from urea and nitrate based fertilizers are well acknowledged (e.g. Harrison & Webb, 2001; EFMA, 2009), a sensitivity analysis was made with urea instead of CAN as the mineral N fertilizer. This affects the calculation of NH_3 and indirect N_2O flows (due to the changed NH_3 emission for the latter), besides inducing an additional CO_2 release. Emission factors considered for estimating NH_3 from urea are shown in Table 4 and CO_2 releases from urea application are estimated as 0.20 kg CO_2-C kg^{-1} urea applied (IPCC, 2006).

Because of its Danish scope, this study considers that crops N requirements are fulfilled by 50% animal manure, and 50% mineral fertilizers. Though this reflects well the conditions of Denmark, a world-leading exporter of pig and dairy products where considerable volumes of manure are available (Dalgaard *et al*., 2011), it does not represent the situation of countries where manure access is limited. Because of the manure consequences on the C and N balances of the crop systems, a sensitivity analysis was carried out to reflect the situation where fertilization is provided by mineral fertilizers only, up to the crop requirements in N, P and K.

A top-down approach has been suggested by Crutzen *et al.* (2008) as an alternative to the IPCC methodology for estimating N_2O emissions, which is based, among others, on global N_2O budgets and on a global budget of 'new' N input to the agricultural system (i.e. synthetic fertilizers and biological nitrogen fixation). Crutzen *et al.* (2008) calculated the ratio between these 'new' N input to agriculture to the global agricultural-related N_2O budget, and the conclusion from this parametrical relationship is that an average of 3–5% of the new reactive agricultural-related N entering the terrestrial biosphere should appear in the atmosphere as N_2O. Because of the important global warming potential of N_2O (298 kg CO_2 equivalent per kg N_2O for a time horizon of 100 years; Forster *et al*., 2007), the methodology proposed by Crutzen *et al.* (2008) is applied as a sensitivity analysis, using the highest factor of the interval (i.e. 5%). In this case, the input by biological N fixation was estimated at 2 kg N ha^{-1} y^{-1}, based on Kristensen *et al.* (2008). As earlier stated, emissions of N_2O generated as a result of the mineralization of soil organic C are also calculated as a sensitivity analysis, based on the IPCC methodology.

Results and Discussion

Life cycle inventory results

Key inventory results are shown in Tables 1–3. As highlighted in Table 1, some crops require a much greater N input than other crops (e.g. ryegrass requires approximately 2.8 times more than spring barley). Table 1 also shows that, except for *Miscanthus* and willow, all crops receive a greater N input on sandy soils, i.e. between 3% and 21%, depending on the crops.

The highest yielding crops in terms of biomass DM are, as illustrated in Table 2, willow, *Miscanthus*, silage maize, sugar beet and ryegrass, *Miscanthus* harvested in autumn being the highest. Table 2 also highlights that it is only for willow and *Miscanthus* (both harvest seasons), whose yields are more sensitive to water availability, that there is a variation between the wet and the dry climate. This is, however, essentially because of a methodological choice. In fact, the data used for annual crops and ryegrass are those from the national fertilization guidelines (Danish Ministry of Food, Agriculture & Fisheries, 2009), as this allowed consistency between yields and N input. Yet, these data do not distinguish between wet and dry climates. However, for most annual crops, there is little effect of dry and wet climate on crop yields, partly because a large share of the difference in rainfall occurs during winter, where it primarily affects N leaching, and partly because some of these crops show relatively little response to rainfall under the climatic conditions of Denmark (Kristensen *et al*., 2011). It is nevertheless acknowledged that different results would be observed in regions with different climatic conditions than Denmark.

Table 2 also shows that yields on sandy loam soils are generally higher than on sandy soils, and this also applies for the above- and below-ground residues. As a result, the C and N input to soil from above- and below-ground residues is generally greater on sandy loam soils for most crops (Table 3).

Soil C balances

On soils with an average initial soil C level, only perennial crop systems as well as winter wheat and spring barley with straw incorporation and catch crop gave rise to an increase in soil C (Fig. 2a). This, however, only applies on sandy soil for the latter, as well as for winter wheat with straw removal and for *Miscanthus* harvested in autumn. This result for winter wheat (without straw incorporation), an annual crop, may appear counter-intuitive, though it simply reflects that wheat is a relatively high yielding crop with a fairly large proportion of biomass that is returned to the soil (Table 2).

With a low initial soil C level, all crop systems led to an increase in soil C (Fig. 2b), while for an initial high soil C level, only three crop systems gave rise to soil C increases. This agrees with previous studies (e.g. Hillier *et al*., 2009) and illustrates how soils with high organic matter content are vulnerable to C losses from cultivation, due to their higher decay rate of organic matter. Losses of soil C are more pronounced on the sandy loam soil (Fig. 2a), which may be due to the lower C:N ratio on this soil compared to sandy soil (10.9 vs. 12.9).

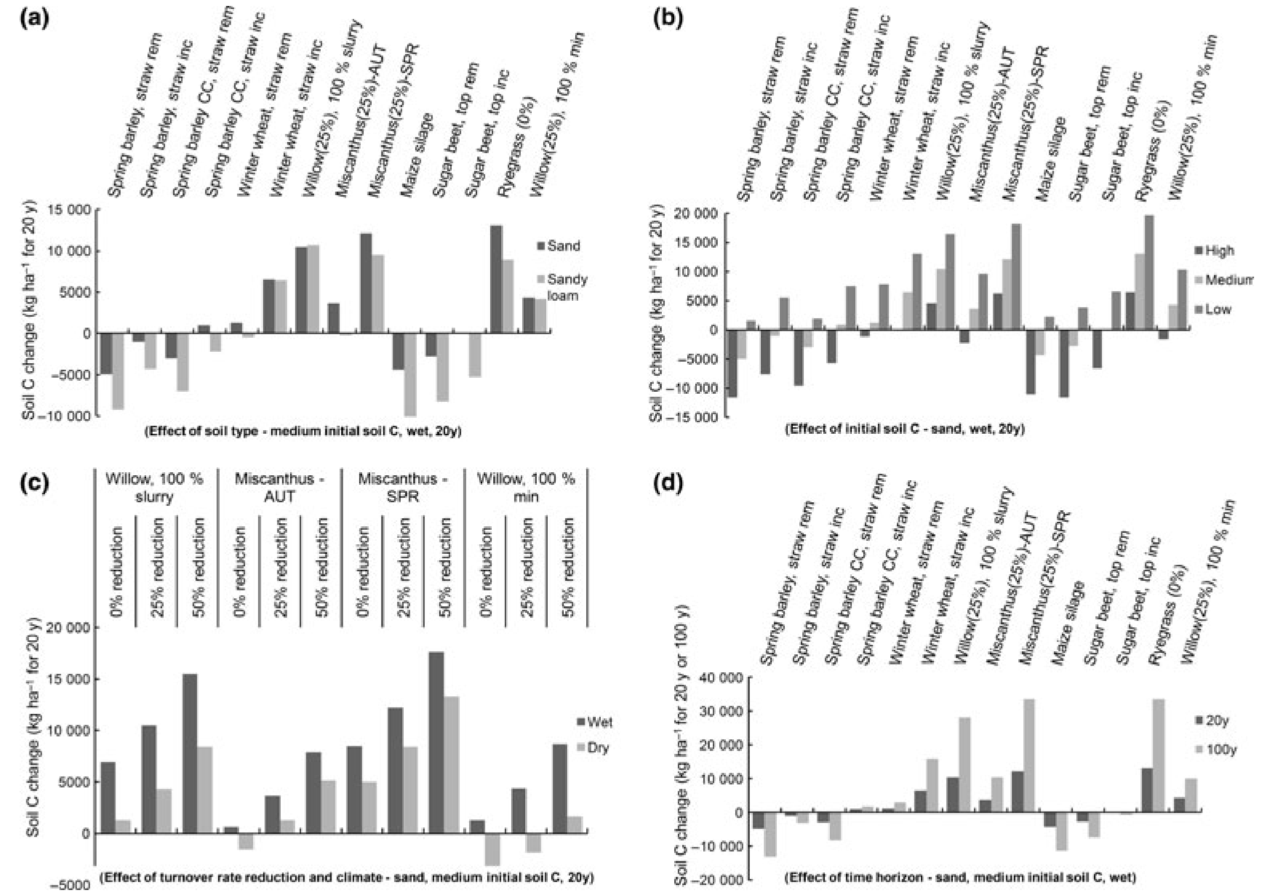

Fig. 2 Effect of soil type (a), initial soil C level (b), climate and turnover reduction rate (c) and horizon time (d) on soil C changes, for all crop systems studied. Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values presented for *Miscanthus* and willow are for established plantations and the value between parentheses indicates the turnover rate reduction considered because of the absence of tillage.

In fact, it has been shown in previous studies that a larger proportion of the soil organic matter is resistant to degradation as the C:N ratio of soils increases (Thomsen *et al.*, 2008).

The effect of climate is only shown for *Miscanthus* and willow as it is only for these crops that this effect is captured (Fig. 2c). Soil C gains are much more important under the wet climate, and this essentially reflects the higher yields under a wet climate (Table 2), where the C input to soil are thus higher.

Reducing the turnover rate in response to the absence of tillage is, as shown in Fig. 2c., important for the soil C results, as the soil C level is at least increased by ca. 45% each time the turnover rate is reduced by 25%. Reductions in soil C turnover rate of 50% have been found for no-tilled soils in Denmark and globally, when considering only the top-soil (Chatskikh *et al.*, 2008, 2009). However, such a large reduction in turnover rate has been questioned when the entire soil profile is considered, and other studies have shown smaller effects of tillage on soil organic matter turnover (Baker *et al.*, 2007; Govaerts *et al.*, 2009). The absence of tillage introduces a completely different depth profile of soil C and also differences in vertical transport of soil C, which may bias estimates of changes in soil C if they are only based on measurements in the top soil. Based on these considerations, the reduction in soil C turnover rate under no-tillage is probably somewhere between 0% and 50%. Given the large importance of this assumption on the soil C changes (Fig. 2c), the use of three different turnover rates for perennial crops in this LCI is therefore very relevant.

Figure 2d presents the overall soil C changes over 20 years vs. 100 years. Results in Fig. 2d are shown for the whole 20 or 100 years, but it may often be needed, when performing environmental assessments, to express these absolute soil C changes occurring over a given time period (here 20 and 100 years) on an annual basis. In order to do so, the IPCC methodology (IPCC, 2006) proposes to divide the changes evenly over each year of the time horizon considered (the IPCC time perspective is 20 years). Because the change in soil C is in fact much more pronounced at the beginning of the period and eventually levels out to reach a new equilibrium (Petersen & Knudsen, 2010), the result of such an annualization is that the annual soil C changes are lower as the horizon time considered is increased, which should be kept in mind when interpreting soil C change figures.

Incorporating straw and tops instead of harvesting them gave a rise in soil C for all crop systems involving a secondary harvest, for a medium initial soil C level, a wet climate, and a horizon time of 20 years (Fig. 2a). For this specific set of conditions, sequestration potentials of straw and tops ranged between 2.7 (beet tops) and 5.2 (wheat straw) Mg C ha^{-1} on sandy soils, and between 3.0 (beet tops) and 6.9 (wheat straw) Mg C ha^{-1} on sandy loam soils (for 20 years). Saffih-Hdadi & Mary (2008), who used their simulation model on data from nine different published studies, presented a similar range of values, i.e. 1.6–7.7 Mg C ha^{-1}, when their results are converted for 20 years.

Also for the abovementioned specific set of conditions, the effect of a catch crop on soil C is not as significant as for straw incorporation, but yet has a non negligible impact on soil C (Fig. 2a), the increase being of approximately 2.1 Mg C ha^{-1}, for 20 years.

Figure 3 presents the C balance for the different crop systems, i.e. the breakdown between the different input and outputs C sources. In Fig. 3a and c, the C outputs are slightly greater than the input for some crop systems. This is because these systems are losing soil C, i.e. some C that was already in the soil is degraded and emitted as CO_2-C (in orange in the output bars). In contrast, Fig. 3b but also Fig. 3a and c show systems where the outputs are slightly lower than the input, the difference reflecting the soil C sequestration that occurred in these systems. Figure 3 illustrates that silage maize, sugar beet and *Miscanthus* (harvested in autumn), are the crops for which the proportion of C ending up as useful yield (blue tones in the output bars) rather than as CO_2 losses (in pink in the output bars) is the highest. In fact, the ratio of the total harvest (primary and secondary yield, when this applies) to the net C

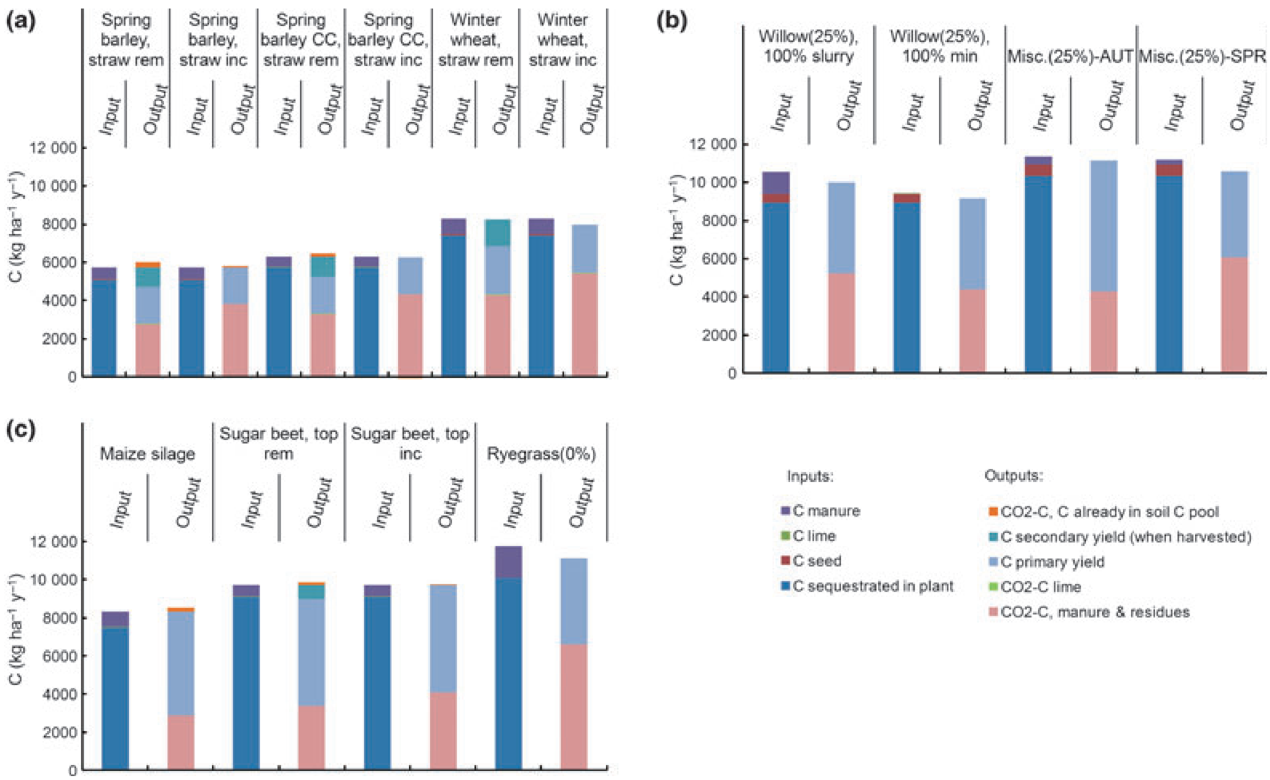

Fig. 3 Breakdown of the biogenic CO_2-C flows for barley and wheat crop systems (a) for Miscanthus and willow crop systems (b) and for silage maize, sugar beet and ryegrass crop systems (c), on sandy soils and under a wet climate, for all systems. Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values presented for *Miscanthus* and willow are for established plantations and the value between parentheses indicates the turnover rate reduction considered because of the absence of tillage.

sequestrated in the plant material (dark blue in the input bars), here referred to as the C turnover efficiency, ranges between 62% and 72% for these crops, while it varies from 33% to 58% for the other crop systems (Appendix S6). To put this into perspective, it illustrates that, for example, 72% of the net C uptake from the atmosphere by maize is found in the harvest, which can be used for energy. On the other hand, only 33% of the net C uptake from the atmosphere by the spring barley and catch crop system (with straw incorporation) is harvested, reflecting that only one-third of the C sequestrated by the biomass in this system can be converted to energy. An important difference between silage maize and sugar beet vs. *Miscanthus* (autumn harvest) crop systems is, however, that these crops contribute to soil C losses (Fig. 2a). Though this aspect appears negligible in terms of C balance (it represents < 4% of the total C outputs), it has consequences for the long-term soil quality, and thus fertility, which are not reflected in a simple C balance (Schjønning *et al.*, 2012).

Overall, higher CO_2-C flows were modelled for the crop systems on sandy loam soils (Appendix S6), which reflects once again the higher yields on this soil type (Table 2), and consequently the higher residue input which has the potential to be emitted as CO_2.

Emission flows of nitrogen compounds

Nitrogen based emission flows are closely related to the amount of N fertilizer that has been applied to the different crop systems (Fig. 4). Ryegrass therefore presents the highest emissions for most N flows, while *Miscanthus* and willow generally present the smallest.

Ammonia emissions are slightly higher (or equal) on sandy soils than on sandy loam soils. This reflects the greater N input on sandy soils compared to sandy loam

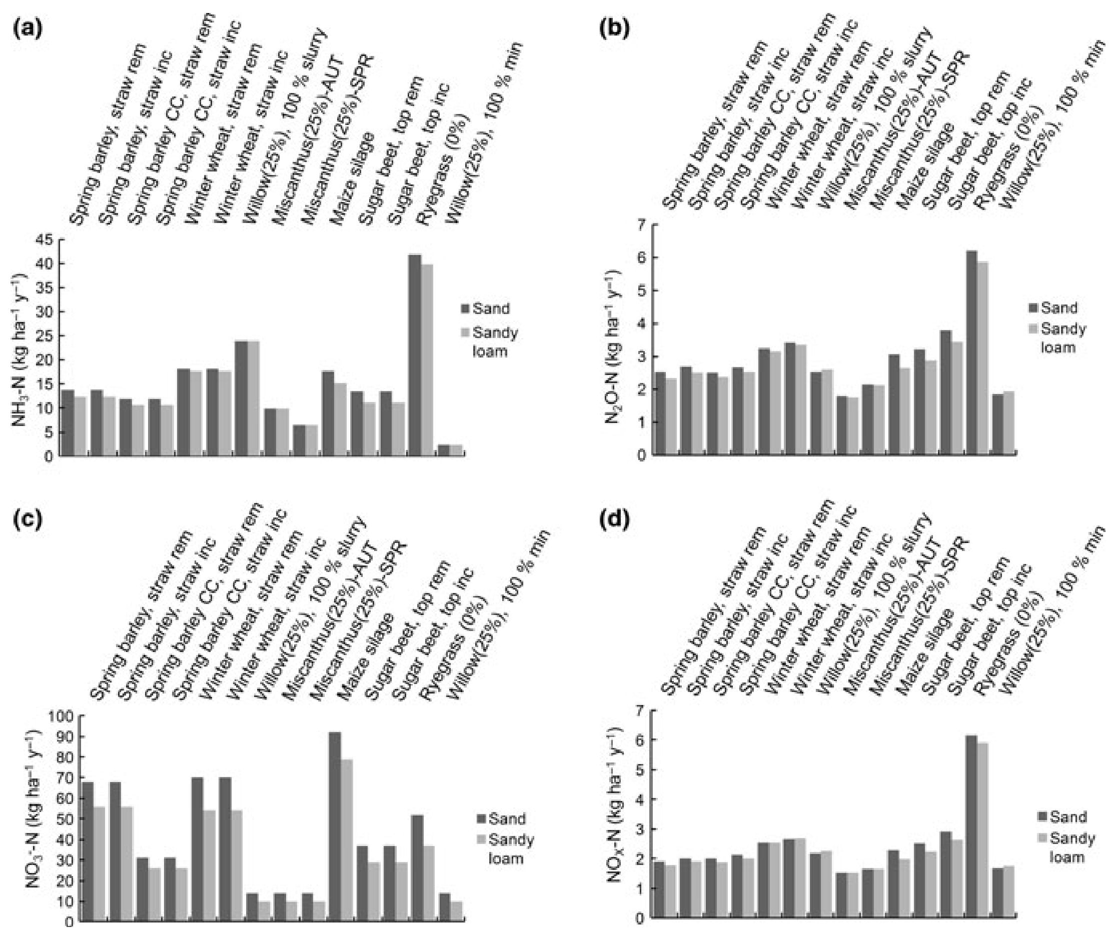

Fig. 4 Emission flows of nitrogen compounds to air and water: NH3 (a), N2O (b), nitrate leaching (c) and NOx (d). Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values presented for *Miscanthus* and willow are for established plantations and the value between parentheses indicates the turnover rate reduction considered because of the absence of tillage. All figures are for a medium initial soil C and a wet climate.

soils. Combining spring barley with a catch crop significantly reduced nitrate leaching (approximately 54% reduction), and to some extent also NH_3 volatilization (approximately 14% reduction). The considerable effect of catch crops on leaching was expected as nitrate leaching reduction is the main purpose of catch crops, and similar magnitudes of NO_3^- reduction have been reported in other studies (Hansen & Djurhuus, 1997; Askegaard *et al.*, 2011). For NH_3, this result is mainly due to the reduced N fertilizer rates used in systems with a catch crop.

Figure 4c shows that NO_3^- results are not affected by straw incorporation. Many studies reported, from a short-term perspective, a decrease in NO_3^- losses with increasing straw incorporation (e.g. Beaudoin *et al.*, 2005; Gabrielle & Gagnaire, 2008), due to a temporary immobilization of mineral N by soil microflora. When the microbes die, this immobilized N is remobilized and as a result, the net effect is simply to postpone the straw-N losses by a few years. For this reason, the empirical model used in this study for predicting nitrate leaching does not consider any effects from the straw incorporation (Kristensen *et al.*, 2008). In the longer term, however, an increase in soil organic matter through incorporation of straw, tops and catch crops may lead to higher levels of NO_3^- leaching than estimated in this study.

The climate type mainly affected the NO_3^- leaching results, and consequently, slightly affected the (indirect) N_2O results as well (Fig. 5).

Other emission flows

Table 5 presents the non C and N flows assessed in this study, for all crop systems, on both sandy and sandy loam soils, for a wet climate. No distinctions were made for the climate type as this only affects *Miscanthus* and willow. Phosphorus, Cu and Zn losses tend to be larger on sandy soils. This reflects the greater surplus of these nutrients applied on sandy soils, due to the larger amount of applied slurry on this soil (which is, under Danish legislation, based on the N requirements of the crops). Incorporating the straw gave rise to increases of P losses varying between 0.12 and 0.22 kg P ha^{-1} y^{-1}. This is due to the greater P input to soil involved when this extra biomass is incorporated. Biogenic NMVOC emitted from photosynthesising leaves of crops are, as expected, larger for perennial than annual crops. It also tends to be greater on sandy loam soil, reflecting the higher crop yields on that soil type (Table 2).

Sensitivity analyses

Sensitivity analyses were performed for the methodology employed to estimate N_2O, as well as for the type of N fertilizer used (CAN vs. urea; system with 50% manure-N vs. no manure), for sandy soil under wet climate (Table 6). The results indicate a general increase of about 46% of NH_3-N when urea is used instead of CAN, though this is much higher for ryegrass and willow (for years where fertilization is performed with 100% mineral fertilizers). In comparison, the effect of the fertilizer type on N_2O-N emissions is quite insignificant, the maximal increase being of 6% (Table 6). These results are consistent with Harrison & Webb (2001), who concluded that replacing urea with nitrate based fertilizers has the potential to significantly reduce NH_3 without affecting N_2O losses. This nevertheless highlights the importance of the type of mineral N fertilizer used on the N flows results, and to some extent on the

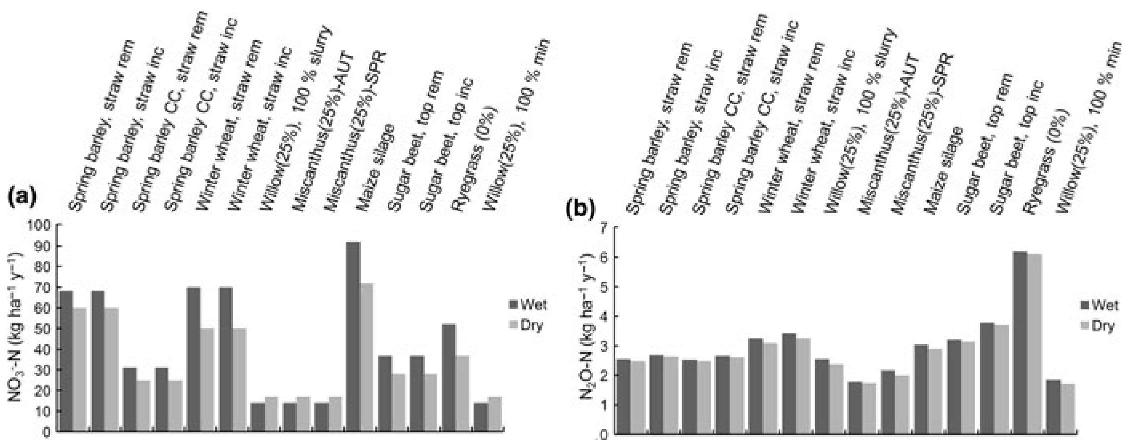

Fig. 5 Modelled effect of climate on nitrate leaching (a) and N2O emissions (b). Straw rem stands for straw removal and straw inc for straw incorporation. CC stands for catch crop, SPR for spring harvest, AUT for autumn harvest and min for mineral fertilizers. Values presented for *Miscanthus* and willow are for established plantations and the value between parentheses indicates the turnover rate reduction considered because of the absence of tillage. All figures are for a medium initial soil C, on sand.

Table 5 Non C and N emission flows resulting of all crop systems, for a wet climate

Crop systems	Straw and tops management*	P Sand	P Sandy loam	NMVOC Sand	NMVOC Sandy loam	Cu Sand	Cu Sandy loam	Zn Sand	Zn Sandy loam
		Nutrient flows (kg ha^{-1} y^{-1}), Soil type							
Spring barley	R	0.36	0.25	0.12	0.14	0.31	0.28	0.42	0.27
	I	0.46	0.37	0.12	0.14	0.32	0.29	0.77	0.66
Spring barley & catch crop	R	0.37	0.27	0.12	0.14	0.27	0.23	0.30	0.15
	I	0.48	0.39	0.12	0.14	0.28	0.24	0.65	0.54
Winter wheat	R	0.18	0.00	0.16	0.20	0.42	0.40	0.90	0.80
	I	0.31	0.15	0.16	0.20	0.43	0.41	1.04	0.97
Willow, 100% slurry	n.a.	0.57	0.53	0.48	0.58	0.57	0.56	1.13	1.01
Miscanthus†, autumn harvest	n.a.	0.15	0.15	0.44	0.44	0.20	0.20	0.18	0.18
Miscanthus†, spring harvest	n.a.	0.23	0.23	0.29	0.29	0.14	0.14	0.13	0.13
Maize silage	n.a.	0.63	0.65	0.00	0.00	0.37	0.31	0.30	0.14
Sugar beet	R	0.87	0.87	0.00	0.00	0.26	0.20	0.00	0.00
	I	1.09	1.09	0.00	0.00	0.28	0.22	0.00	0.00
Ryegrass	n.a.	0.27	0.31	0.45	0.40	0.84	0.80	2.04	1.97
Willow, 100% mineral	n.a.	0.16	0.12	0.48	0.58	0.00	0.00	0.00	0.00

*R: straw or top removal; I: straw or top incorporation; n.a: not applicable.
†For established *Miscanthus*.

C flows, because of the CO_2 emissions induced by urea application. The sensitivity analysis considering that no manure is applied showed similar tendencies, i.e. a considerable effect on NH_3 emissions, and a smaller effect on N_2O, the effect being in the same order of magnitude for all crops (Table 6). This reflects that these bioenergy systems, when cultivated in countries where manure is not as available as in Denmark, would present the same relative performance with respect to the N flows. Of greater interest is the impact of zero manure input on soil C changes. The soil C losses with no manure input become much greater, and the gains much reduced. Further, some of the systems that presented soil C gains with manure application now present soil C losses (spring barley & catch crop with straw incorporation; winter wheat with straw removal). This highlights, for winter wheat, that straw removal will be sustainable (soil C wise) only if manure is applied. The soil C balance for spring harvested *Miscanthus* appears less affected by this change. This reflects the considerable input of C from above- and below-ground residues for this crop, as compared to other crops (Table 3). Since this inventory considers no input of mineral Cu and Zn, not applying manure would mean no Cu and Zn losses. For P, it is only for ryegrass that the losses would differ, since it is the only crop (together with willow 100% manure), where no mineral fertilizers needed to be added to the manure input in order to fulfil the crop P requirements (Appendix S8). In this case, the P losses of the ryegrass system would be reduced from 0.27 to 0.05 kg P ha^{-1} y^{-1}. For NO_3^- leaching, ryegrass is also the system where the losses would be the most reduced (ca. 19%) by a zero manure input (Table 6). For the other crop systems, where much less manure-N is applied, the effect is much more limited, being below 10% reduction of NO_3^- leaching.

The top-down approach developed by Crutzen *et al.* (2008) for estimating N_2O emissions yielded greater N_2O emission estimates for most crops, except for sugar beet, spring harvested *Miscanthus*, and willow in years where the fertilization is ensured by 100% animal slurry (Table 6). For the latter case, the important difference obtained reflects the fact that the methodology of Crutzen *et al.* (2008) accounts only for the 'new' N, and thereby does not reflect the N that is constantly recycled, e.g. the N from manure or from crop residues. In a system where N fertilization is provided by animal manure only, resulting N_2O estimates are in consequence likely to be underestimated. It should be highlighted that the N_2O estimates derived from the Crutzen *et al.* methodology are based on the highest factor (i.e. 5%) suggested by Crutzen *et al.* (2008). Interestingly, if the lower factor of 3% would have been used, the N_2O results from the Crutzen *et al.* methodology would have been lower than from the methodology used in this study (based on IPCC) by 16–98% (except for willow, 100% mineral fertilizers). The results of this sensitivity analysis highlight the importance of the methodology used to model N_2O emissions, which can

Table 6 Sensitivity analysis results. All values are in kg ha^{-1} y^{-1}, unless otherwise indicated, for a sandy soil, under wet climate. Eventual inconsistencies are due to rounding

Substance flow affected	Parameter tested	Spring barley (R)*	Spring barley (I)*	Spring barley & catch crop (R)*	Spring barley & catch crop (I)*	Winter wheat (R)*	Winter wheat (I)*	Willow (100% slurry)	Miscanthus (autumn) Year 4–20	Miscanthus (spring) Year 4–20	Maize silage	Sugar beet (R)*	Sugar beet (I)*	Ryegrass	Willow (100% mineral)
NH_3-N, total	Urea	20.1	20.1	17.4	17.4	26.5	26.5	23.9	14.4	9.6	25.8	19.6	19.6	75.2	14.4
	CAN	13.8	13.8	11.9	11.9	18.2	18.2	23.9	9.9	6.6	17.7	13.5	13.5	41.9	2.4
	No manure†	2.5	2.5	2.2	2.2	3.3	3.3	-	1.8	1.2	3.2	2.5	2.5	5.1	2.4
	Δ fertlizer‡	46%	46%	46%	46%	46%	46%	0%	46%	46%	46%	46%	46%	80%	>100%
	Δ manure§	−82%	−82%	−82%	−82%	−82%	−82%	-	−82%	−82%	−82%	−82%	−82%	−88%	0%
N_2O-N, total	Urea	2.6	2.8	2.6	2.7	3.3	3.5	2.5	1.8	2.2	3.1	3.3	3.8	6.5	2.0
	CAN	2.5	2.7	2.5	2.7	3.3	3.4	2.5	1.8	2.2	3.1	3.2	3.8	6.2	1.9
	No manure†	2.2	2.3	2.2	2.4	2.8	2.9	-	1.5	2.0	2.6	2.9	3.4	5.2	1.9
	SON¶	2.7	2.7	2.6	2.7	3.3	3.4	2.5	1.8	2.2	3.2	3.3	3.8	6.2	1.9
	Crutzen *et al.* method	3.3	3.3	2.8	2.8	4.3	4.3	0.1	2.4	1.6	4.2	3.2	3.2	8.7	6.1
	Δ fertlizer‡	2%	2%	2%	2%	3%	2%	0%	3%	1%	3%	2%	2%	5%	6%
	Δ manure§	−14%	−13%	−12%	−12%	−14%	−14%	-	−14%	−8%	−15%	−11%	−9%	−17%	0%
	Δ SON**	7.5%	1.5%	4.6%	0%	0%	0%	0%	0%	0%	5.5%	3.3%	0%	0%	0%
	Δ method††	28%	21%	12%	6%	31%	24%	−96%	31%	−26%	36%	−1%	−16%	40%	>100%
CO_2-C	Induced from urea	27.0	27.0	23.4	23.4	35.6	35.6	0.0	19.3	12.9	34.7	26.4	26.4	73.3	51.4
Soil C changes‡‡	Manure§§	−248	−51	−149	48	65	326	524	183	609	−218	−138	−0.5	656	218
	No manure	−397	−200	−270	−81	−132	128	-	73	534	−411	−283	−146	249	218
	Δ manure§	−60%	<−100%	−81%	<−100%	<−100%	−61%	-	−60%	−12%	−89%	<−100%	<−100%	−62%	0%
N leaching	Manure§§	68	68	31	31	70	70	14	14	14	92	37	37	52	14
	No manure	64	64	29	29	65	65	-	14	14	85	34	34	42	14
	Δ manure§	−6%	−6%	−6%	−6%	−7%	−7%	-	0%	0%	−8%	−8%	−8%	−19%	0%

*R: straw or top removal; I: straw or top incorporation.
†This sensitivity analysis considers that crops are fertilized with 100% CAN.
‡Illustrates how much, in %, the results would be higher (or lower) with urea instead of CAN.
§Illustrates how much, in %, the results would be higher (or lower) if crops would be fertilized with 100% CAN (i.e. no manure) instead of being fertilized with 50% animal manure, and 50% CAN.
¶Soil organic nitrogen. Values presented in this line are the total N_2O emissions (with CAN as a fertilizer) when the emissions stemming from mineralization of soil organic matter (when there are losses of soil C), are also included, based on the IPCC methodology (IPCC, 2006).
**Illustrates how much, in %, the results would be higher (or lower) if the N_2O from the mineralization of soil organic C, when there are losses of soil C, would be included.
††Illustrates how much, in %, the results would be higher (or lower) with the methodology from Crutzen *et al.* instead of the methodology employed in this study (with CAN as a fertilizer).
‡‡Annualized over 20 years. The minus sings indicate a loss of SOC.
§§This corresponds to the baseline scenario where 50% CAN, and 50% animal manure is applied. For willow, however, it is only 100% CAN or 100% manure.

considerably affect the global warming potential result (in CO_2 eq.) of a given crop system. The increase in N_2O obtained, if the N_2O resulting from the mineralization of soil organic C is accounted for, based on the IPCC methodology (IPCC, 2006), varies between 0% (systems without soil C losses) and 7.5% (Table 6), illustrating that accounting for this effect or not have a limited impact on the N_2O balance.

Limitations and applicability

An important limitation of the study relates to the quality of the data used for the inventory. This applies particularly for the above- and below-ground residues estimates, which are of tremendous importance when modelling the soil C changes. For example, the approach used in this study for modelling below-ground residues assumed a fixed ratio between above- and below-ground biomass. Though this approach is commonly used in inventories of soil carbon balances in agricultural systems (Johnson *et al*., 2006), recent experimental evidences cast doubt on this assumption. In reality, the below-ground input may be much less dependent on above-ground biomass (Chirinda *et al*., 2012), and this would lead to an overestimation of the below-ground carbon input in the present study. In this light, the availability of long-term field measurements for above- and below-ground residues would surely increase the quality of bioenergy LCAs. It should also be highlighted that the yields used in this study for willow and *Miscanthus* are based on very few experiments compared to the extensive data on the other crops considered in this study, and yields of perennial crops may increase as insight is gained on the optimal management practices for these crops. However, even though the absolute values of each individual parameter considered in this LCI could be improved, the relative differences modelled between each crop systems are believed to reflect the reality observed in Northern European fields.

This inventory was built to reflect the specific situation of Denmark. This applies, for example, with respect to the fertilization rates used, the soil types chosen, the crop yields and the use of manure (though a sensitivity analysis without manure application was also performed). However, the inventory, because of its high disaggregation and transparency, can easily be adapted, with the methodologies presented in this study, so it can as well be used for assessing bioenergy systems of other regions. Moreover, the results presented in this inventory may be used directly as a proxy to reflect the situation of neighbouring European countries, where fertilization rates are similar to those presented in this study.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Appendix S1. *Miscanthus* and willow life cycles.

Appendix S2. Modeling details for the 9 main agricultural operations involved: soil preparation, propagation, liming, sowing, plant protection, fertilization, irrigation, harvest and transport field-farm.

Appendix S3. Primary yields data details for willow and *Miscanthus*.

Appendix S4. Estimates for above- and below-ground residues of perennial crops.

Appendix S5. N leaching estimates for *Miscanthus* and willow.

Appendix S6. Carbon balance.

Appendix S7. Calculation of NMVOC from photosynthesizing leaves.

Appendix S8. Calculation of phosphorus, cooper and zinc losses.

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Paper II

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Bioenergy Production from Perennial Energy Crops: A Consequential LCA of 12 Bioenergy Scenarios including Land Use Changes

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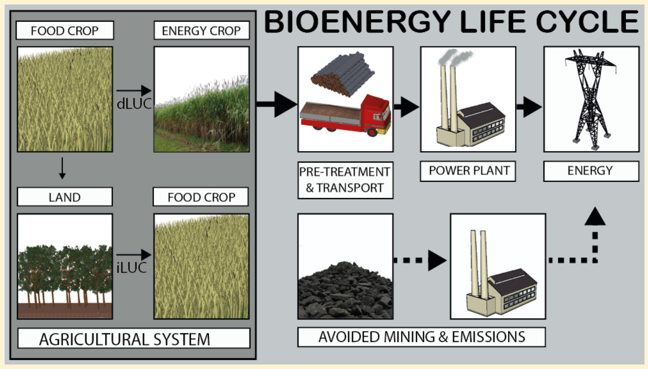
Supporting Information



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

1. INTRODUCTION

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

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

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

The goal of this study is to assess the environmental impacts associated with the production of bioenergy (heat and electricity) from 1 ha (10,000 m^2) of Danish arable land cultivated with ryegrass, willow and *Miscanthus*, considering four different biomass-to-energy (BtE) conversion pathways: (i) anaerobic co-digestion with manure, (ii) gasification, (iii)

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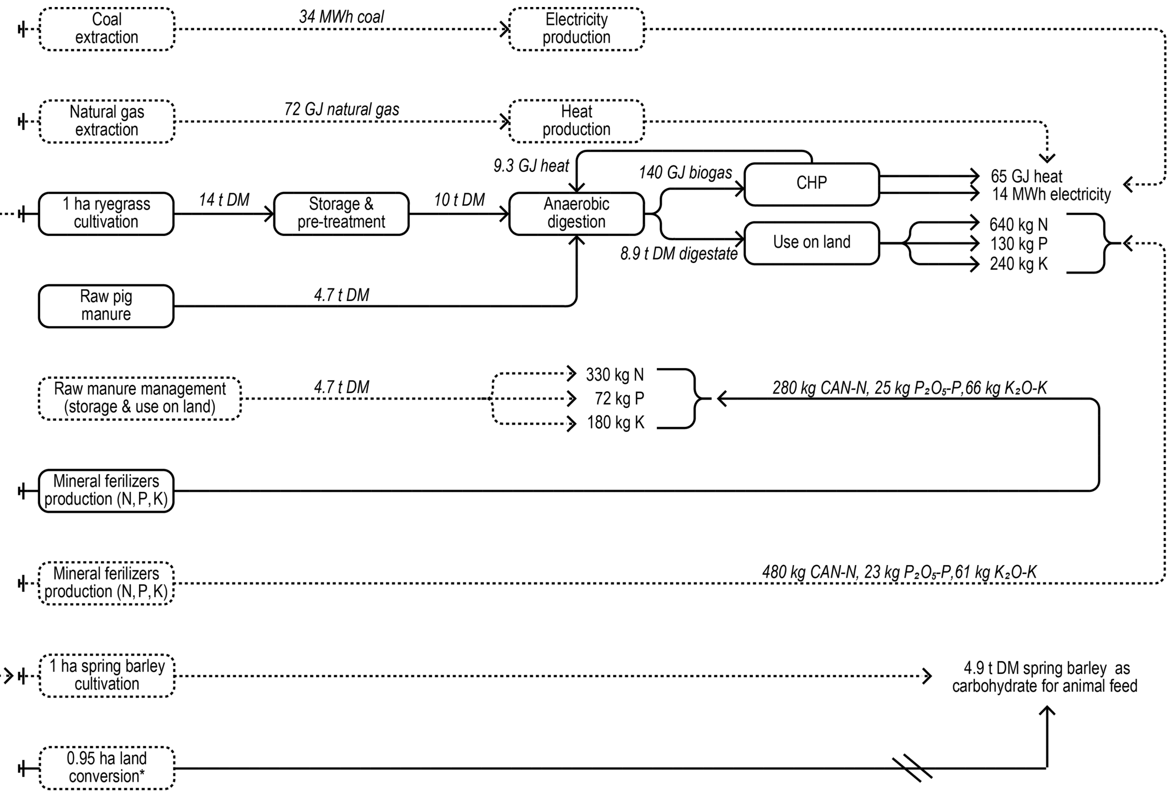

Figure 1. Process flow diagram for the bioenergy scenario anaerobic co-digestion of ryegrass with raw pig manure. Electricity and heat produced represent net values (i.e., plants own consumptions have been subtracted). (*) Not all the converted land is to be cultivated in barley, and not all the Danish barley displaced is replaced, due to various market mechanisms. Values rounded (2 significant digits).

combustion in small-to-medium scale biomass combined heat and power (CHP) plants and iv) co-firing in large scale coal-fired CHP plants.

2. MATERIALS AND METHODS

2.1. Life Cycle Assessment Model. *2.1.1. Scope and Functional Unit.* The environmental assessment presented in this study was performed using consequential life cycle assessment (LCA).[15,16] The functional unit upon which all input and output flows were expressed was 1 ha of agricultural land used to grow the selected energy crops. The geographical scope considered for the LCA was Denmark, that is, the data inventory for crops cultivation and BtE plants were specific for Danish conditions. Similarly, the legislative context of Denmark (e.g., fertilization) was considered. The temporal scope considered was 20 years, i.e., all assessed systems were operated for 20y duration.

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

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

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

As illustrated in Figure 1, the digestate produced from anaerobic digestion was used as a fertilizer (for N, P, and K), which avoided marginal mineral N, P, and K fertilizers to be produced and used, based on the content of N, P, and K of the digestate. The marginal N, P, and K fertilizers considered were calcium ammonium nitrate, diammonium phosphate, and potassium chloride, respectively, conformingly with refs 14

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and 24. Further, based on the model from ref 24, it was considered that the manure portion used for co-digestion would have otherwise been stored and applied on land, without digestion or other treatment.

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

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

2.3. Life Cycle Inventory (LCI). *2.3.1. Crops.* The LCI of all crops was based on a recent Danish consequential LCI,[14] which comprises all processes involved during the cultivation stage, up to harvest. This included the tillage activities, liming, propagation (seed, rhizome, and cutting production), plant protection, fertilization, sowing/planting, harvest, and transport from farm to field. A sandy loam soil has been considered for all crops, as well as precipitations of 964 mm y^{-1}. For *Miscanthus* and willow, the C turnover rate in the topsoil was considered to be reduced by 25% in response to the absence of tillage over many years. For all crops, the fertilization operations were performed in conformity with Danish regulations,[33,34] involving an upper limit for the amount of N to be applied on the field, both as mineral fertilizer and animal slurry.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

3. RESULTS AND DISCUSSION

3.1. Carbon and Nitrogen Flows. The induced C and N flows for ryegrass, willow and *Miscanthus* are presented in Figures S13–S18 (SI).

As illustrated in SI Figures S13–S15, more than 85% of the C input to the energy crop system (the most notable being the uptake from the atmosphere) ends up emitted as CO_2, whether as a result of the cultivation stage or as a result of the final energy use. As indicated in refs 8,44, many bioenergy studies report rather different results, as the biogenic CO_2 emissions from the cultivation stage (releases from manure and residues not entering the soil C pool), which here represents 50–57% (SI Figures S13-S15) of the C input fate, are not accounted for. This highlights the importance of the error made if a complete system-based mass balance, such as the one performed in this study, is not considered.

The C from atmospheric uptake was similar for all the three crops (about 11–12 t C ha^{-1} y^{-1}): for all crops, only about half of this C ended up in the harvested biomass, the other half ending up in the non-harvested above- and below-ground residues (SI Figures S13–S15). The biogenic CO_2 emission related to crop cultivation (6.1 to 6.9 t CO_2–C ha^{-1} y^{-1}) was also in the same order of magnitude for all crops (SI Figure S13–S15; Table S8). The biogenic carbon emission from the final energy use, however, varied significantly more (2.9 to 6.0 t CO_2–C ha^{-1} y^{-1}), as detailed in SI Table S8. This reflects the importance of two main parameters: the crop yield and the BtE technology. In fact, the biogenic CO_2 emission from the final energy use was the greatest for thermal treatments (combustion and gasification), where 95–100% of the carbon was emitted as CO_2, whereas it was significantly lower for biological treatment (anaerobic co-digestion), where only ca. 40–46% of the crop (and raw manure) carbon was gasified (SI Table S8). This unconverted C during anaerobic co-digestion is ultimately applied on land, through the digestate. However, this did not represent a significant carbon sink, as more than two-thirds of this C was released as CO_2, rather than sequestrated in the soil (SI Figure S13–S15). This is in accordance with previous findings (e.g., ref 24).

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

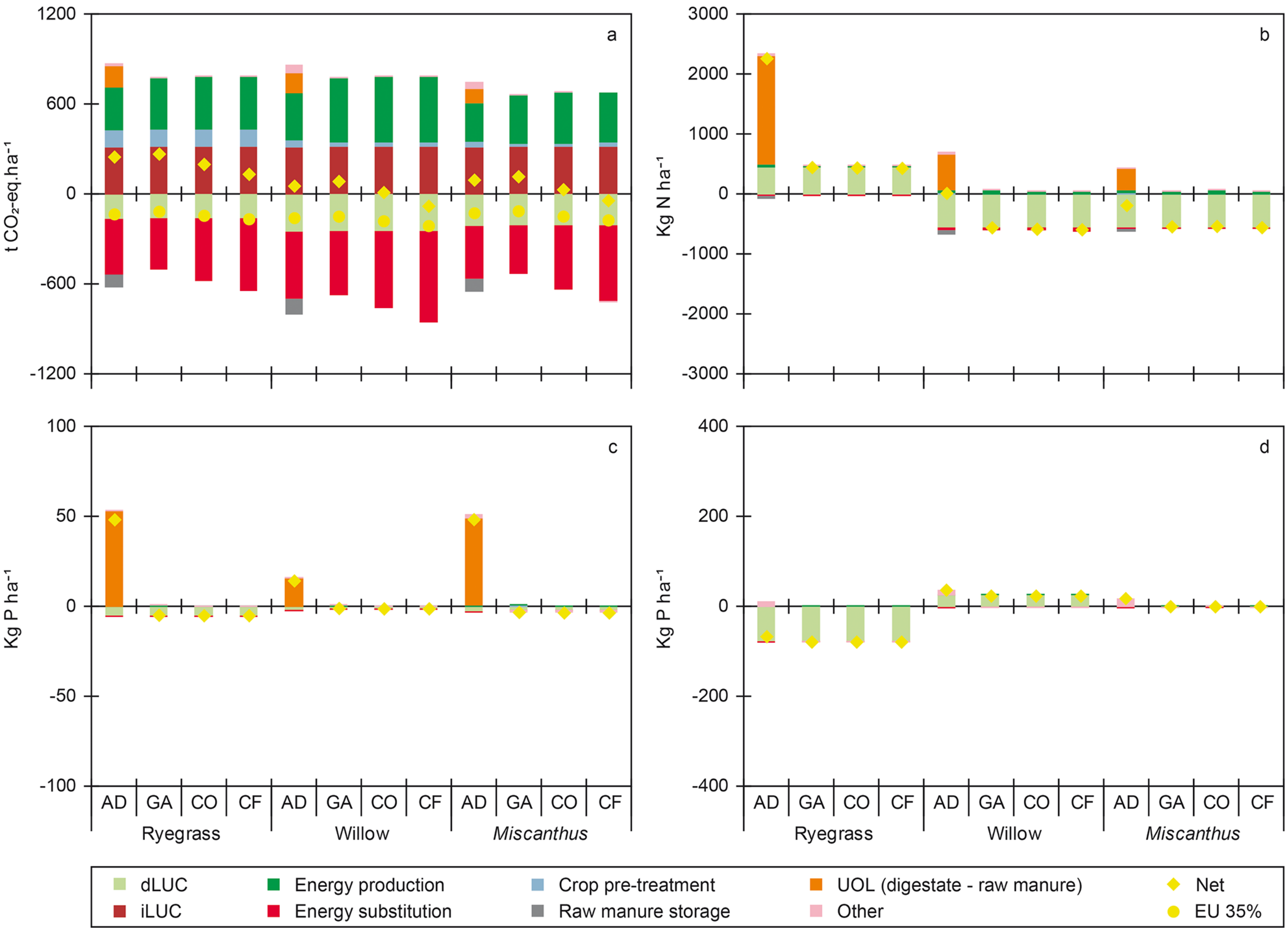

Figure 2. LCA results for (a) global warming (over a 100 year horizon, t CO_2-eq. ha^{-1}); (b) aquatic N-eutrophication (kg N ha^{-1}); c) aquatic P-eutrophication (kg P ha^{-1}); and d) phosphorus as resource (kg P ha^{-1}). All systems represent a 20 year time scope.

digestion of *Miscanthus* with raw pig manure) to 8.3 (co-firing of willow) t C ha^{-1} y^{-1} (SI Table S8).

As opposed to C, the outputs of N flows were more diversified among the individual flows. The most significant N flows occurred during the UOL of the digestate for the anaerobic co-digestion scenarios, and during the cultivation stage for the other scenarios (SI Figures S16–S18; Table S8). Ryegrass showed the highest emissions of N during the cultivation phase; these occurred as a consequence of the higher nitrogen fertilizer requirements of ryegrass (450 kg N ha^{-1} y^{-1}) compared with willow (170 kg N ha^{-1} y^{-1}) and *Miscanthus* (71 kg N ha^{-1} y^{-1}). These fertilization rates (and the related N-based emissions) are based on today's practices, but should be seen as reflecting the highest end of the interval. In fact, *Miscanthus* and willow are relatively new crops, and it can be expected that lower application rates will be required as insight is gained on the optimal management of these crops.[45,46] Similarly, lower N application could be considered for ryegrass dedicated to bioenergy, where protein production is not the focus (as in the case of forage ryegrass). The N-related emissions at the UOL stage (anaerobic co-digestion scenarios) were similar for all the three crops, as a consequence of the Danish legislation for fertilization fixing the maximal amount of N to be applied in agricultural fields.[33,34] Overall, NO_3^- and NH_3 emissions were the most significant N-emissions.

3.2. Indirect Land Use Changes. The iLUC impacts of the studied bioenergy systems were the same for all scenarios (Figure 2a), as they all had the same "point of origin": the conversion of 1 ha of Danish land (cultivated with spring barley) to energy crops. As shown in Table 1 (and further detailed in the SI), these iLUC impacts were estimated to 310 t CO_2-eq. ha^{-1} (±170 t CO_2-eq. ha^{-1}). The impacts were annualized over a period of 20 years in accordance with IPCC[47] and with prominent European legislation,[48] corresponding to about 16 t CO_2-eq. ha^{-1} y^{-1} (or 70–130 g CO_2-eq. MJ, calculated dividing the annualized iLUC impact by the energy introduced into CHP units each year, dry basis).

Although currently debated and relatively uncertain,[49] the iLUC impact quantified here can contribute with important learnings: (i) it is not zero; and (ii) it may cover a significant proportion of the overall global warming impact (Figure 2a) (between one-third and half of the positive contributions, depending on the scenario), and cancels out the otherwise avoided GHG emissions in the scenarios. Moreover, it should be highlighted that the 310 t CO_2-eq. ha^{-1} obtained here only covers the GHG related to the net expansion resulting from the modeling of ref 29 and does not include the GHG related to the intensification of crop production (which accounts, based on the results of (29), to about 30% of the displacement response). This suggests that the "real" impact may actually be higher. The only other LCA study[50] the authors were aware of

Table 1. Estimation of the iLUC CO_2 Impact[a]

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

[a]Eventual inconsistencies due to rounding (numbers are reported with 2 significant digits). [b]Indicated biomes are as in ref 29. When the biomes mentioned in ref 29 did not figure in the biomes from the Woods Hole Research Centre data,[9] an equivalent was considered, which is indicated between parentheses, when it applies. [c]Based on the results from ref 29. [d]With xss: Sub-Saharan Africa, excluding Botswana, Lesotho, Namibia, South Africa and Swaziland; xeu15: EU-15, excluding Denmark; bra: Brazil; xsu: Former Soviet Union, excluding the Baltic States; aus: Australia; can: Canada; xla: South America, excluding Brazil and Peru; usa: United States. As indicated in ref 29 this aggregation covers 92% of the total net expansion. [e]From the Woods Hole Research Centre, as published in ref 9. [f]Considering that 25% of the C in soil is converted, for all biomes, except when forest is converted to grassland, where 0% of soil C is converted; 100% of the C in vegetation is converted for all forest biomes; 100% of the C in vegetation is converted for tropical grasslands; 0% of the C in vegetation is converted for all other biomes. [g]The conversion per ha is made considering that it is 1 ha of spring barley that is initially displaced, with a yield of 4.9 t DM ha^{-1} and a DM content of 85% of the crop fresh matter, based on ref 14. [φ]The maximal and minimal range are based on the qualitative description of the uncertainty related to the biomes conversion results made by ref 29. The levels identified as "very good", "good" and "moderate" were considered as an uncertainty of ±20%, 40%, and 60%, respectively.

attempting to quantify iLUC on the basis of an hectare of land displaced (and not a biofuel mandate shock) led to a considerably higher value, that is, 440−560 t CO_2-eq. ha^{-1} (considering a 20 years period and only conversion of forest). Although it cannot be directly compared, our annualized iLUC value (70−130 g CO_2-eq. MJ^{-1}, calculated dividing the annualized iLUC impact by the energy introduced into CHP units each year, dry basis) lies within the range of values found in ref 10 for marginal increases in the demand for biofuels.

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

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

On the selected impact categories, global warming appears critical as only two scenarios indicate overall savings for this category compared with the fossil fuel reference. Only co-firing of willow and *Miscanthus* indicated net overall savings, that is, these were the only two scenarios for which an environmental benefit, GHG-wise, was identified in relation to using 1 ha of land for bioenergy. However, the magnitude of the global warming impacts found in this study (between −82 and 270 t CO_2-eq. ha^{-1} over 20 years) was much higher than previous results from literature. For instance, ref 54 calculated a saving between ca. −18 and −35 t CO_2-eq. ha^{-1} y^{-1} (ca. −360 to −700 t CO_2-eq. ha^{-1} over 20 years) for bioenergy systems based on willow and *Miscanthus* plantations in Ireland;[55] quantified savings about −25 t CO_2-eq. ha^{-1} y^{-1} (about −500 t CO_2-eq. ha^{-1} in 20 years) for bioenergy systems based on *Miscanthus* plantations in Italy;[56] estimated a saving between ca. −10 and −11 t CO_2-eq. ha^{-1} y^{-1} (about −210 to −220 t CO_2-eq. ha^{-1} in 20 years) for *Miscanthus* and willow plantations in the UK. The reason for these differences is that this study, as opposed to the previous, considered iLUC, which has tremendous significance on the overall GHG balance as earlier discussed.

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

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

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

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

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

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

ASSOCIATED CONTENT

Supporting Information

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

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Notes

The authors declare no competing financial interest.

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Paper III

Methodological aspects of environmental assessment of livestock production by LCA (Life Cycle Assessment)

Methodische Aspekte der Umweltbewertung tierischer Erzeugung mit Ökobilanzen

Lorie Hamelin, Henrik Wenzel

Summary

This paper illustrates the necessity to use a holistic perspective when striving to assess the environmental performance of a livestock production system. It elaborates on the methodological dimension of livestock- related LCAs, i.e. it describes the essential environmental impacts categories to include, the stages of the livestock system to comprise and the effects on adjoining systems like energy and fertilizer production to account for. Further, it explains the comparative nature of environmental assessment and how to use reference systems as the basis of comparison of alternative techniques. It illustrates a Danish example of establishing data of such reference systems. Finally, it provides an overview of approaches used to estimate emissions based on knowledge of manure composition throughout the stages of the livestock production system.

Zusammenfassung

Dieser Artikel verdeutlicht die Notwendigkeit einer ganzheitlichen Betrachtungsweise bei der Bewertung der Umweltwirkungen von Tierhaltungssystemen. Die Studie arbeitet die methodische Dimension der Ökobilanzierung in der Nutztierhaltung heraus. Beschrieben werden die unbedingt zu berücksichtigenden Umweltwirkungskategorien, die einzubeziehenden Abschnitte von Tierhaltungssystemen und die zu berücksichtigenden Wirkungen auf benachbarte Systeme wie Energieerzeugung und Düngerproduktion. Darüber hinaus erklärt die Studie die vergleichende Eigenschaft der ökologischen Bewertung und beschreibt, wie Referenzsysteme als Grundlage für den Vergleich alternativer Vorgehensweisen zu verwenden sind. An einem Beispiel aus Dänemark wird erläutert, wie die Daten aus solchen Referenzsystemen ermittelt werden. Abschließend wird ein Überblick über Methoden der Emissionsabschätzung gegeben, die auf der Kenntnis der Wirtschaftsdüngerzusammensetzung in allen Bereichen eines Tierhaltungssystems basieren.

1 The dimensions of the livestock LCA

Mitigation technologies for the livestock production system endeavour to reduce the emissions to the environment from this sector of activities. However, reducing the emissions from livestock production most often has a dual effect. While applying a technique that reduces emissions from e.g. housing systems, the manure composition may well change as a result. If for example ammonia emissions are reduced from housing, the nitrogen content of the manure increases. As subsequent downstream emissions often depend on the manure composition, these emissions will change as well. Moreover, as the fertilizer value of manure changes, so does its ability to replace mineral fertilizers. Such dual – or multiple – effects call for a whole-system assessment when assessing the environmental consequences of new techniques.

The necessity of a whole-system approach as a basis for assessing the environmental impacts from livestock production is widely recognized among both scientists and decision makers within policymaking and management.

The decision maker's concern to have a robust platform for the decision is, for example, a high priority concern in the context of assessing and nominating techniques as "best available techniques" (BAT) under the EU Industrial Emissions Directive (EC 2010). It is not a wishful situation to nominate a technique as BAT, if unforeseen environmental side-effects later emerge and call for a re-evaluation and removal of the technique from the BAT category. This may contribute to undermining the credibility of the BAT approach to environmental administration.

Even though whole-system considerations are comprehensive by nature, the required knowledge is typically available among experts within the various parts of the system. The time invested in pinpointing all possible environmental consequences related to the application of a given technique may be well spent compared to what has to be invested in dealing with justified critique afterwards.

In the context of assessing livestock production, three essential issues justify a system approach:

1. the need to include the whole spectrum of substances affected;
2. the need to consider the whole chain of production and;
3. the need to consider relations with adjoining systems and related consequences.

These three dimensions will be further elaborated in the following.

2 The need to include all substances affected

Livestock production involves many processes of biogenic nature where implemented techniques may impact several substance flows simultaneously, particularly for nitrogen (N) and carbon (C) flows. The microbial processes taking place in manure and soil are diverse and involve the transformation of substances in the spectrum from organic nitrogen to various forms like ammonia/ammonium (NH_3/NH_4^+), nitrous oxide (N_2O) and nitrate (NO_3^-) and from organic carbon to carbon dioxide (CO_2) and methane (CH_4). As a result, acting on one targeted flow has simultaneous consequences on another flow.

To illustrate these interdependences, the technique of acidifying slurry from in-house slurry pits can be used as an example. This technique (i.e. lowering slurry pH) can be applied to reduce the emission of ammonia from animal houses, as in this example, but also from outdoor storage and/or field. However, while a low pH reduces NH_3 emissions by pushing the equilibrium towards ammonium (NH_4^+), it also inhibits the CH_4 producing methanogenic bacteria, which results in a reduction of CH_4 emissions as well. In this case, accounting for NH_3 only would not capture this additional benefit and would not provide a fair assessment of the full potential of the technique.

In the example of slurry acidification, the described interdependencies implied further benefits. But such interdependences may also reveal trade-offs between environmental advantages and disadvantages. For instance, an air cleaning technique using a biofilter/bioscrubber to treat emissions from housing units could be very efficient at reducing ammonia and odours, but may also contribute to the unintended emission of dinitrogen oxide (N_2O) generated from the microbial processes in the filter. Dinitrogen oxide is a greenhouse gas 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), meaning that even small releases can have a large impact on the overall greenhouse gas balance of the technique. This implication would be missed out if only the substances targeted by the technique are measured and/or studied, in this case ammonia and odours. Such a case illustrates the potential implications (e.g. wrongly labelling a technique as BAT) of not considering all the changed flows resulting from the application of the technique.

3 The need to include the whole chain

The livestock production chain may be summarized as four main system stages, at which a technology can be applied: 1) feed and feeding systems, 2) housing systems including in-house manure management and storage, 3) outdoor manure management and storage, and 4) field application of manure. This is illustrated in Figure 1.

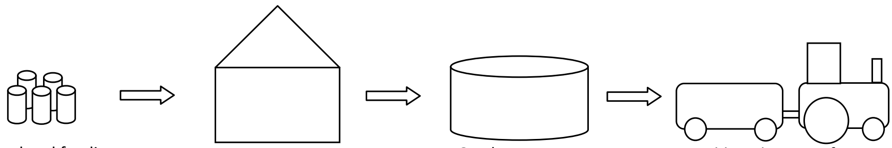

Fig. 1: The 4 main system stages of livestock production

Any intervention in the livestock production system by applying a new technique at any stage of the system may influence environmental impacts from the whole chain, typically from the point of application and downstream the chain. In some cases, an intervention in the system may even influence environmental aspects upstream the point of application, though this is less often the case.

The effects downstream the point of application appears rather obvious when emission flows are closely related to the manure composition. The composition of manure, in particular in N and C, is the starting point for estimating the major emission flows from it like ammonia, methane or nitrous oxide by various recognized methodologies and models. Therefore, keeping track of changes in manure composition from one system stage to another is a way to estimate the related emission flows.

Effects upstream the point of application of a technique are less typical, but still relevant. Examples are changes in feed or diet where the production of feed constituents upstream is avoided or changed. Reducing e. g. soy protein addition to the feed by specific amino acid addition can give rise to very large reduction in upstream greenhouse gas emissions from the soybean production.

Such downstream and upstream system aspects are illustrated below by 3 examples.

Example 1: acidification of slurry in pig houses

The first example refers to the case of slurry acidification in slurry pits mentioned also earlier. This case illustrates the situation where the technique has an impact at the point of application and downstream the chain of production. As mentioned before, the acidification of slurry in-house affects NH_3 and CH_4 flows. Consequently, CO_2 flows are also affected, and as indicated in a recent LCA study (Wesnæs et al. 2009), N_2O emission is likely to be reduced as well. Acidification, thus, results in an overall reduction of these emissions from the animal houses compared to the case without acidification. The same benefits are also seen for the subsequent outdoor storage (Wesnæs et al. 2009). Moreover, the benefits of acidified slurry comprise reduced NH_3 emissions even further downstream, i. e. from field application of the slurry. However, when emissions from the slurry are reduced, the remaining nitrogen content of the slurry is of course increased.

The higher N content of acidified slurry combined with conditions favouring N_2O emissions (e.g. soil in transition between aerobic and anaerobic conditions), thus, may lead to higher N_2O emissions than what these emissions would have been without the slurry acidification. Moreover, application of acidified slurry to fields is likely to lead to an increased need for application of lime. On the other hand, the increased ammonium N content of the slurry leads to an increased amount of crop available N, potentially resulting in higher crop yield and/or higher mineral fertilizer replacement, all other parameters remaining equal.

As this example illustrates, only considering the consequences on the emission flows at the point of application (i.e. housing unit) does not allow to capture the benefits of the technique during outdoor storage and on the field and thereby does not provide a fair assessment of the technique. Similarly, the potential drawbacks occurring at the field stage (increased lime application and potential N_2O increases) would also not be reflected by considering only the point of application.

Example 2: peat biofilter

A second example consists of a biofilter using peat as a bed media, used as a mitigation measure for odour emissions from manure storage (in-house and/or outdoor storage). This example is an illustration of a situation where the technique has impacts at the point of application and upstream the chain of production. At the point of application, odours, NH_3 and potentially also hydrogen sulphide (H_2S) are reduced as compared to the situation without the application of the peat filter. As this technique does not influence the manure composition (as e.g. in the acidification case), it does not interfere with subsequent manure handling. Consequently, emissions are not changed downstream the chain. However, the implementation of the technique involves peat mining, an activity giving rise to very significant greenhouse gas emissions (Parish et al. 2008). These emissions may prove to be significant relative to other greenhouse gas emissions in the livestock system.

Example 3: manure separation

Manure separation can provide many environmental benefits both at the point of application (e.g. reduced NH_3 formation and emission deriving from reduced contact between urea of urine and urease enzyme from faeces) and downstream the chain (e.g. in the field by favouring a better nutrient management). However, the on-farm management of both liquid and solid fraction is crucial for the overall benefits of the technique. For example, if the solid fraction is left in aerobic conditions allowing composting to occur (e.g. stored in heap without cover), the pH, temperature and decomposition of organic matter in the manure will increase, which results in significant N losses, mostly through

NH_3 but also N_2O, as well as C losses as CO_2 and CH_4. These losses, as illustrated by several field studies (Amon et al. 2006, Dinuccio et al. 2008, Hansen et al. 2006), may result in manure separation to cause an overall increase in greenhouse gas and ammonia emissions, compared to the case without separation.

4 The need to consider interactions with adjoining systems

The previous sections illustrated that 1) the implementation of a technique may simultaneously change the flows of several substances and 2) that such changes may also have impacts both downstream and upstream the point of application of the technique.

The implementation of a technique, however, does not only affect the emission flows from the livestock production system itself, but also those of other systems with which the livestock system is coupled. In fact, environmentally significant processes that are not necessary part of the livestock system may be induced or displaced as a result of applying the technique in the livestock production system.

The most significant environmental implications of a new technique or managerial procedure may indeed often be found within such relations to adjoining systems. Typically involved adjoining systems resulting from an intervention in the livestock system are the energy production system, the fertilizer production system, the feed production system (organic and inorganic ingredients), as well as the meat and crop production system themselves. This is illustrated in Table 1.

Tab. 1: Generic examples of interactions between the livestock production system and key adjoining systems resulting from a technological intervention in the livestock system

	Type of intervention	Possible consequences	Affected adjoining system
(a)	Any intervention allowing energy production or savings	saving or substitution of fossil energy	energy production
(b)	Any intervention changing the nutrient content and/or availability in manure	substitution of mineral fertilizer	fertilizer production
		change in crop yield	crop production
(c)	Any intervention changing the nutritional value and/or composition of the feed	a change in feed production, both from the organic (e.g. crops) and inorganic (e.g. phosphate) ingredients	feed production
		a change in feed conversion (i.e. kg feed crop per kg animal weight)	meat production

The types of interventions (a), (b) and (c) of Table 1 are further illustrated with specific examples.

Example 1: Intervention affecting the energy production system (type a)

An obvious illustration of the application of a technique affecting the energy production system is the example of biogas production. When biogas is produced from the digestion of slurry, this biogas may be burned in a biogas engine, thereby producing both heat and power. The power produced can be used through the national electricity grid, thus substituting another fuel, typically a fossil fuel (e.g. coal, natural gas). Avoiding this fossil fuel based electricity production (and the environmental impacts related to it) is a direct interaction between the livestock system and the energy system. Similarly, the surplus heat produced with the biogas (i.e. the net heat remaining after one part has been used for the needs of the system itself) may be used in the district heating system or for domestic heating at the farm itself, thereby avoiding another fuel (e.g. natural gas). Instead of being burned in a biogas engine, the biogas of this example could also have been upgraded and injected into the natural gas grid, in which case it would interact with the energy system by directly substituting natural gas.

Further, the anaerobic digestion of the slurry also results in the production of digested slurry with enhanced efficiency as nitrogen fertilizer, as a result of the conversion of organic nitrogen to ammonia nitrogen having higher N availability to the crops. Yet, the potentially increased crop yield induced by the digested slurry involves interactions with the crop production system, since this extra amount of crop produced will replace similar crops on the market. Consequently, the environmental impacts from such crop production are also avoided and this consequence should be considered in the assessment.

The use of the digested slurry for fertilization is also in itself an interaction with the mineral fertilizer production system. This is further elaborated in the next example.

Example 2: Intervention affecting the fertilizer and/or crop production system (type b)

The in-house slurry acidification example described before can be used to highlight this type of intervention, i.e. where the fertilizer and the crop production systems are affected. In this case, even a third adjoining system is also influenced.

Because the acidified slurry has a higher N content than untreated slurry (due to the N that is not lost through volatilisation), a higher amount of plant available N is applied to the field. This can lead to two types of consequences, either higher crop yields due to the higher fertilizer value, or to increased replacement of mineral fertilizer. The overall consequence may include both. The amount of extra crop then produced does not have to be produced somewhere else, which means that the environmental impacts from such production are avoided. The same holds for the extra mineral fertilizer avoided. Moreover, the added sulphur (S) of the acidified slurry (i.e. the S from the sulphuric acid) can replace a portion of the mineral S fertilizer to be applied (S is, like N, P and K, an essential plant macronutrient). This synthetic S consequently does not need to be produced and the environmental impacts related to this production are then avoided – balancing to some extent the production of sulphuric acid for the acidification in the first place.

An additional adjoining system involved in this acidification example consists of the extra lime that may need to be applied to the field in order to ensure a proper soil pH. Without the intervention of the acidification technique, this extra lime would not have been produced (and used).

Example 3: Intervention affecting the feed and/or meat production system (type c)

This type of intervention is exemplified by the technique of adding phytase to pig or poultry feed. Because the phosphorus contained within phytate, the principal storage form of phosphorus in feed ingredients of plant origin, cannot be readily digested by monogastric animals like poultry and pigs, inorganic phosphorus is supplemented in order to meet the animal nutritional needs. However, when phytase is added in the diet, it releases the phosphorus from feed phytate, so it becomes available for digestion, thus reducing the phosphorus required from supplements. This is, thus, a direct interaction with the production of inorganic phosphorus for feed. Because less phosphorus will resist digestion, the manure composition will change as well, its overall phosphorus content being reduced (because more is absorbed by the animals). This may then involve interactions with the fertilizer production adjoining system as it may (or may not) displace application and production of mineral P fertilizer, depending on the existing P balance of the field in question and the applying environmental regulations on fertilization.

This particular case may even involve interactions with the meat production system. In fact, phytase may also contribute to increase the overall amount of digested amino acids by the animal as it has been shown to act on the release of some phytate-bounded amino acids that would typically be resistant to digestion. As a result, the growth of animals is affected and thus the meat production system. Based on this particular case of phytase addition in feed, Figure 2 illustrates the induced interactions with adjoining systems throughout the whole livestock production system.

In summary, assessing the impacts of a technique at the point of application only, provides incomplete information about the technique, and will lead to insufficiently informed and sometimes wrong judgements on the environmental implications of the technique. It is only through a qualified and to the extent necessary a quantified assessment of all impacts occurring throughout the whole system and adjoining systems that reliable conclusions can be drawn about the overall environmental performance of a technique.

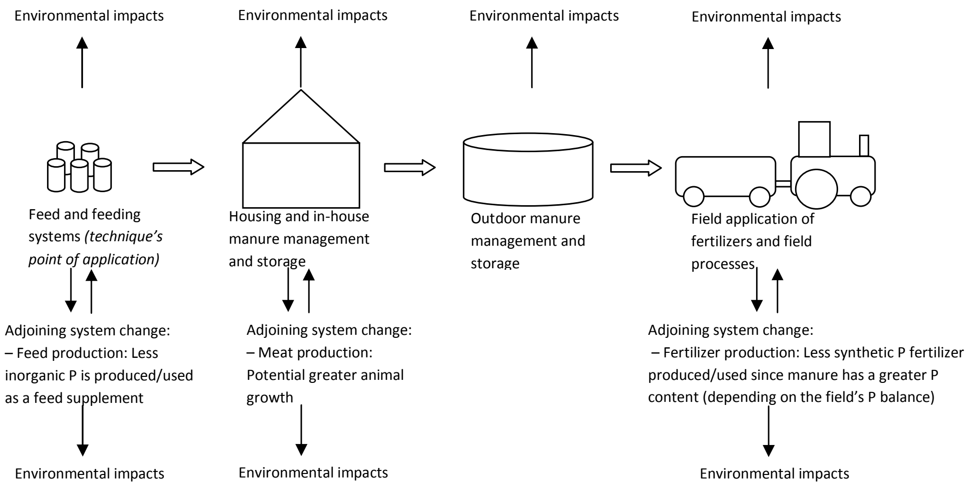

Fig. 2: Illustration of the interactions between the IRPP system and its adjoining systems in the case of phytase addition to the feed

5 The essential impacts categories in the livestock LCA

The major environmental consequences induced by techniques to be implemented in livestock production system will typically involve the following main environmental impacts:

- Global warming: mainly through changes of CH_4, CO_2 and N_2O flows
- Acidification: mainly through changes of NH_3 flows, and less considerably also H_2S and nitrogen oxides (NO_x)
- Nutrient enrichment: mainly through changes of nitrogen and phosphorus flows
- Odour: mainly through changes of flows of volatile organic compound (VOC), sulphur-containing compounds (e.g. H_2S and mercaptanes) and nitrogen-containing compounds (e.g. ammonia and amines)
- Fine particles (PM_{10} and $PM_{2.5}$): e.g. through any changes in emissions of dust and aerosols
- Toxic substances: to reflect the implications of toxic substances accumulating in the environment that the use of some techniques could entail
- Other: any impacts of special relevance to the technique to be assessed

The set of impact categories should be chosen as part of the assessment process for the technique in question in order to cover the environmentally most relevant aspects of the technique.

It can be argued to include "ammonia" as one category of its own instead of e. g. a category named "acidification". This is because ammonia generally is, for livestock systems, a substance of great environmental interest, not only due to its acidifying properties but also other impacts like toxicity from aerosols on humans.

The category "greenhouse gases" also includes CO_2, a greenhouse gas typically not considered as a concern in environmental assessment of animal production systems because CO_2 emissions from livestock production are of a biogenic nature. However, some techniques can induce changes in the biogenic CO_2 flows in a magnitude making significant differences in the greenhouse gas balance of the technique (versus the reference), and to account for this, CO_2 has to be included. An example is the differences in land requirements, as the use of marginal land throughout the world can imply large CO_2 emissions from land cultivation. Arable land has the particularity of being a finite and thus constrained resource, which involves that competition arises for its various uses (e. g. food, feed, providing feedstock for bioenergy or for the green chemistry sector, providing shelter). Because of that, techniques impacting the amount of land used for the livestock system (e. g. through changes in yields, through legislation when the implementation of some techniques allows for extending the production, through a change in the feed system) imply, as a final consequence, repercussions on how the land is used worldwide (e. g. expansion of arable land at the expense of nature). These implications on land are comprehensively documented and typically referred to as "land use changes". Such land use changes inevitably involve significant changes in releases of soil C to the atmosphere, in magnitudes depending of the land types being converted.

6 Scope definition and data

The assessment must be relative, i. e. reflect the changes compared to a reference. This is argued by the fact that any environmental assessment must by definition be comparative, i. e. the implications of a given technique lie in the changes it induces as compared to a reference situation. Moreover, a relative accounting will allow references to be different and thus provide a balanced evaluation of the environmental performances of a technique between the various regions and contextual conditions throughout e. g. the EU.

Identifying the reference system

There are differences in the prevailing reference systems around EU and, therefore, variations in what is the reference. This is partly due to differences in technological state-of-the-art and managerial practices partly due to climatic differences. It is, thus, difficult to identify one common EU reference as background for the assessment, and it is not called for either.

The purpose of the reference is partly to serve as a representative of a ”common” conventional practice. But the main purpose is, however, to serve as a measure-stick to compare and quantify alternative techniques against. This will ensure a common ground for the assessment and quantification. The reference should ideally represent an average or some fair representation of a sound conventional livestock production. But it is not essential to the assessment exactly what level or percentile of environmental performance the reference represents. As long as it is a well known and common reference, and as long as it is used as a common background for all compared techniques, it serves its function.

An effort should be made to simplify the variety of references and create a few operable references suitable as background for the assessment of new techniques. In the effort to simplify, one should consider the two main components of the reference separately:

- The managerial reference, i.e. the technological status and managerial practices of the production system. These are likely to differ from country to country.
- The environmental impact reference, e.g. emissions and other impacts deriving from the technological reference system.

In the decision on which references to include, one should consider which variables are necessary to fix. For example, one might choose to distinguish between cattle, pigs and poultry in the first place and subsequently between sows with suckling pigs and fattening pigs. More variables to consider are: liquid versus solid manure, type of floors, covered/not covered storage, field application technique, type of soil, etc. In this process it may be chosen either to include several variants or to select only one base assumption and then include variations in a discussion of sensitivity.

Example of a Danish reference system for fattening pigs

The following example of a reference system is taken from a recent Danish project which aim is to establish a foundation for life cycle assessment of pig and cow slurry management technologies in Denmark (Wesnæs et al. 2009).

Managerial reference

This fattening pig reference example does not pretend to represent an average of all slurry management systems in Denmark, but was rather defined in order to reflect the “typical” manure management practices in Denmark. Accordingly, an attempt to identify the most commonly used methods and practices has been made, in collaboration with different national experts in the animal rearing area.

As a starting point, it was decided to base the reference upon "fattening pigs" (30–100 kg) and the slurry deriving from this. According to Dalgaard (2007), the majority of farms in Denmark have exclusively fattening pigs. Based on Statistics Denmark (2010), fattening pigs represented, from 1998 to 2010, about 70 % of the total Danish pig holdings (categories "weaners under 50 kg" plus "pigs for slaughtering, 50 kg").

For defining the reference slurry management in this Danish example, it has also been necessary to define some preconditions regarding e.g. housing units, type of storage, field spreading techniques and reference cropping scenarios. The main preconditions that needed to be defined are described below:

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 a national expert (Poulsen 2008). This assumption was necessary because of the reference used for determining the manure composition. A storage time in the pit of approximately 14 days is assumed (Sommer et al. 2009).

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

Outdoor storage: In Denmark, it is required by law to cover outdoor slurry storages in order to reduce ammonia emissions and odour. 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 (Rasmussen et al. 2001).

Transport distance from storage to application to fields: Based on different Danish studies (Rasmussen and 2003, Jacobsen et al. 2002), 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 DLBR (2005), 68 % of all slurry was spread by trail hose application tanker in Denmark (in 2004), and this is still the most common method today (personal communication with Birkmose (2008) and with Pedersen (2008)). Therefore, it is considered that pig slurry is applied with trail hose tankers to the field in the reference scenario.

Soil types: Relevant soil types for pig production and application of pig slurry are clay soil and sandy soil. According to Halberg and Nielsen (2003), pig farms on clay soil constituted 29 % of the total Danish pig meat production in 1999, and pig farms on sandy soil constituted 49 % of the total Danish pig meat production. Accordingly, the reference scenario is set up for both clay soil and sandy soil for pig slurry spreading.

Crop rotation: A crop rotation was defined for assessing nutrients uptake and reference emissions from the field. In collaboration with national experts from the Faculty of Agricultural Science (Aarhus University), a 6-years crop rotation was defined for fields receiving the pig slurry to be applied, with slurry N (kg/(ha • y) 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).

Reference manure composition

The reference slurry for this project was determined based on the Danish normative system for assessing slurry composition (Poulsen et al. 2001, DJF 2008). Poulsen et al. (2001) established the technical background report, and the yearly updated values are published by DJF (2008). These data are combined with data from the literature and mass balances are performed to assess the slurry composition after the different losses.

The composition of the slurry in the reference scenario is calculated at three points:

- Slurry “ex-animal”, i.e. right after excretion
- Slurry “ex-housing”, i.e. in the slurry pit under the animals right before flushing to the pre-tank
- Slurry “ex-storage”, i.e. after months of covered outdoor storage, and right before application to field

For N, P and K, the starting point for determining the slurry composition was the content of these in the slurry ex-animal, which was given in DJF (2008). The amount of these nutrients in the slurry ex-housing and ex-storage was then determined based on an estimation of the losses occurring during in-house and outdoor storage, respectively. On the other hand, the starting point for the dry matter (DM), carbon (C), cooper (Cu) and zinc (Zn) content was the amount of these in the slurry ex-storage, based on the data from the literature. Similarly as for N, P and K, losses during storage (in-house and outdoor) needed to be estimated so the composition of the slurry ex-housing and ex-animal could be estimated.

The reference slurry composition considered in this Danish reference example is presented in Table 2.

Tab. 2: Reference pig slurry composition for the Danish reference example

Parameter	Unit	Slurry ex-animal	Slurry ex-housing	Slurry ex-storage	Data source and assumptions
Slurry	t/t slurry ex-animal	1	1	1.086	considering a water addition of 86 kg during outdoor storage.
Total nitrogen (N)	kg/t slurry	6.60	5.48	4.80	N ex-animal values are from DJF (2008). Values for slurry ex-housing and ex-animal are based on losses during housing and during storage (NH_3, N_2O, N_2, NO) and on dilution. See Table 3 for details about N losses.
Phosphorus (P)	kg/t slurry	1.13	1.13	1.04	P ex-animal values are from DJF (2008). No losses are considered during housing and storage (only dilution).
Potassium (K)	kg/t slurry	2.85	2.85	2.60	K ex-animal values are from DJF (2008). No losses are considered during housing and storage (only dilution).
Dry matter (DM)	kg/t slurry	77.4	69.7	61.0	DM ex-storage values are from DJF (2008). Losses during storage: 5 % of the ex-housing values; losses during housing: 10 % of the ex-animal value. Assumptions for losses during storage and housing are based on Poulsen et al. (2001).
Volatile solids (VS)	kg/t slurry	64.2	56.5	48.8	VS are assumed to constitute 80 % of the DM content of slurry. 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).
Carbon (C)	kg/t slurry	37.0	33.3	29.2	C ex-storage = 47.9 % of DM ex-storage, based on the ratio C: DM obtained by Knudsen and Birkmose (2005). Losses of C during storage and housing assumed to follow the same pattern as DM (i.e. 5 % of the ex-housing values and 10 % of the ex-animal values).
Copper (Cu)	kg/t slurry	30.0	30.0	27.6	Cu ex-storage = 0.0453 % of DM ex-storage, based on the ratio C: DM obtained by Knudsen and Birkmose (2005). No losses considered during housing and storage (only dilution).
Zinc (Zn)	kg/t slurry	89.4	89.4	82.4	Zn ex-storage = 0.135 % of DM ex-storage, based on the ratio C: DM obtained by Knudsen and Birkmose (2005). No losses considered during housing and storage (only dilution).

Tab. 3: Assumptions for N losses in the establishment of slurry composition

Loss	Data source and assumptions
NH_3-N	in-house: 16 % of N ex-animal (Poulsen et al. 2001) outdoor storage: 2 % of N ex-housing (Poulsen et al. 2001), the N ex-housing being estimated according to Poulsen et al. (2001), i.e.: N ex-animal – NH_3-N losses in-house (and not accounting for other losses)
N_2O-N	in-house: 0.002 kg N_2O-N per kg N ex-animal (IPCC 2006) outdoor storage: 0.005 kg N_2O-N per kg N ex-animal (IPCC 2006)
N_2-N	in-house and outdoor storage: assumption that N_2-N = N_2O-N • 3 (based on data from Dämmgen and Hutchings 2008)
NO-N	in-house and outdoor storage: assumption that NO-N = N_2O-N • 1 (based on data from Dämmgen and Hutchings 2008)

Environmental impact reference

The environmental impact reference related to this example will not be described here, but is available in Wesnæs et al. (2009), Annex A.

Getting the data for the assessment

The outset of the data inventory is the scope of the system as determined – adjusted through subsequent iterations. The request for data comprises:

Foreground data:

- Data on reference scenarios (not specific to the technique in question)
- Data on performance of the technique in question at the point of application
- Data on any upstream and/or downstream influences of the technique on processes within the livestock system

Background data:

- Data on any influences on adjoining systems

Foreground data

The source of data for reference scenarios and specific techniques are e.g. available measurements, relevant scientific literature, existing LCAs, and recognized estimation methodologies (e.g. IPCC). Table 4 present an overview of some of the most used approaches to estimate emissions based on manure composition data.

Tab. 4: Targeted emission flows for establishing the reference farm systems and a draft overview of estimation approaches. The establishment of an operational guide for estimation methods is currently on-going.

Emission	Housing	Manure storage	Manure land-spreading
Odour	May be estimated based on regional studies	May be estimated based on regional studies	May be estimated based on regional studies
NH_3-N	May be estimated as a % of the N in ex-animal manure.	May be estimated as a % of the N in ex-housing manure.	May be estimated based on regional studies.
N_2O-N	May be estimated based on IPPC methodology (IPPC, 2006, table 10.21 and table 11.1).	May be estimated based on IPPC methodology (IPPC 2006, table 10.21 and table 11.1).	May be estimated based on IPPC methodology (IPPC 2006, table 11.1 and 11.3).
N leaching	Leaching, if any, can be determined based on regional studies.	Leaching, if any, can be determined based on regional studies.	May be estimated based on regional studies, on empirical models, or on nitrogen balance that considers crop uptake, denitrification, soil organic N changes and ammonia losses.
NO_x-N[a)]	May be estimated from NO-N data, if no data exist on NO_2-N. Can also be derived from a ratio NO : N_2O emissions obtained from relevant scientific publications	May be estimated from NO-N data, if no data exist on NO_2-N. Can also be derived from a ratio NO : N_2O emissions obtained from relevant scientific publications	Most likely not needed for the present purpose. Else can be estimated as for housing and storage. Can also be based on Nemecek and Kägi (2007), p. 36
N_2-N	May be derived from a ratio N_2 : N_2O emissions obtained from relevant scientific publications	May be derived from a ratio N_2 : N_2O emissions obtained from relevant scientific publications	Most likely not needed for the present purpose. Else can be estimated as for housing and storage
CH_4	IPCC (2006) Tier 2: CH_4 [kg] = VS [kg] × B_0[b)] × 0.67 [kg CH_4/m³ CH_4] × MCF[c)], with the ex animal VS	IPCC (2006) Tier 2: as for in-house storage, but with the appropriate MCF. Note: the VS to be used is the ex animal one	Can be considered as negligible under aerobic field conditions
CO_2	If data on manure composition are determined for each stage, may need to be calculated in order to close the mass balance. Else, biogenic CO_2 from manure can be calculated as a function of the CH_4, based on the Buswell equation (Symons and Buswell 1933)	If data on manure composition are determined for each stage, may need to be calculated in order to close the mass balance. Else, biogenic CO_2 from manure can be calculated as a function of the CH_4, based on the Buswell equation (Symons and Buswell 1933)	Biogenic CO_2 emissions due to soil C changes under the given manure application and crop rotation may be estimated from dynamic modelling, if possible

Continue next page

Emission	Housing	Manure storage	Manure land-spreading
P losses	May apply or not. If it applies, it can be determined based on regional studies.	May apply or not. If it applies, it can be determined based on regional studies.	Can be calculated as a % of surpluses, the % value being determined by the ratio P losses: P surpluses reported for the geographical area. Can also be calculated as a % of the P applied (Hauschild and Potting 2005, annex 6.3).

a) NO_x corresponds to the sum of NO and NO_2.
b) B_0: maximum methane producing capacity for the given manure type. Given in IPCC (2006), table 10A4 to 10A9.
c) MCF: methane conversion factor (%). The MCF factor is defined in the revised IPCC (IPCC 1997) guidelines in chapter 4 (on page 4.9) as follows: "Methane Conversion Factor (MCF): The MCF defines the portion of the methane producing potential (Bo) that is achieved. The MCF varies with the manner in which the manure is managed and the climate, and can theoretically range from 0 to 100 per cent. Manure managed as a liquid under hot conditions promotes methane formation and emissions. These manure management conditions have high MCFs, of 65 to 90 per cent. Manure managed as dry material in cold climates does not readily produce methane, and consequently has an MCF of about 1 per cent. Laboratory measurements were used to estimate MCFs for the major manure management techniques." In (IPCC 2006), default MCF values are presented in Table 10.17 for different manure management system and in function of the average annual temperature.

The downstream effects of a technique are often due to the change in slurry/manure composition caused by applying the technique. When this is the case, generic models of emissions from storage and field application of the manure/slurry can often be used to quantitatively estimate emissions. Experts on housing, storage and field application systems have access to such models.

Background data

The variety of data to gather on adjoining systems is relatively limited, as mentioned before, and concerns mainly data on electricity, heat, fertilizer production, feed, crops and animal production. In the LCA community, a large effort is already undertaken to provide such databases, and the effort of extracting the required data for the purpose of LCA-based assessments of potential BAT is ongoing and will be quite small.

Handling data gaps

Whenever data are lacking, a qualified estimate for judging the significance of the data gap is better than just leaving the gap. Data gaps may be filled in by "expert judgements", but it should be emphasized that expert judgements means knowledge-based judgements of what can be quantified and what not. Rather leave an uncertainty open and qualify the proportion of the uncertainty than filling the knowledge gap with a non-substantiated guess.

Standardizing the units

All input flows and emissions flows need to be expressed on the basis of a standardized unit in order to make the reference farm usable. It is here recommended to express data per unit of functional output, i. e. per kg of meat produced or per head for a given year. For techniques that only affect the manure and not the animal production, it is equally valid to express data per ton of excreted manure (i. e. manure "ex-animal"). In order to be able to make use of data expressed in other units, a translation standard should be developed, and data expressed e. g. per animal head or animal place and/or per time unit should be translated per kg or per ton of manure.

7 Concluding remarks

Doing LCAs on livestock systems is a precondition for assessing their environmental performance. The special characteristics of livestock systems are that any intervention or change in the system influences not only the emissions at the point of intervention, but most often also the composition of the manure. As downstream emissions are directly related to the manure composition, it is not meaningful to assess the environmental consequences at the point of intervention only. More and more data on livestock system emissions become available and so do better methods for expressing emissions related to manure composition and contextual conditions.

At present, available estimation approaches are judged to be sufficiently good to allow for a meaningful holistic assessment (LCA) of livestock systems, and for sure better than omitting the holistic perspective as such. The comprehensiveness of the holistic perspective is not necessarily more time consuming, it just implies focusing the available time on the environmental issues that matter the most. It is always better to be approximately right than precisely wrong.

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Paper IV

Environmental Science & Technology

ARTICLE

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Environmental Consequences of Future Biogas Technologies Based on Separated Slurry

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S Supporting Information

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 cosubstrates. 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 bypassing 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's. When designed and operated properly, ensuring, for example, 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 equiv), 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 cosubstrate. However, the availability of applicable organic residues is rather limited compared to slurry volumes. In, for example Denmark, around 5% of the slurry goes through a biogas plant with codigestion 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 cosubstrate 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

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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 (SI). The study is set up in a way allowing for future comparisons with any alternative slurry management techniques.

■ MATERIALS AND 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 refs 13 and 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", that is, 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, for example, 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 SI. The temporal scope is 30 years, based on the lifetime 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), that is, 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 SI.

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, that is, postanimal, posthousing (as it leaves the in-house storage) and poststorage (as it leaves the outdoor storage). Additional details about the reference slurry are available in the SI.

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 unseparated 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, that is, 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 SI.

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 fiber 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.

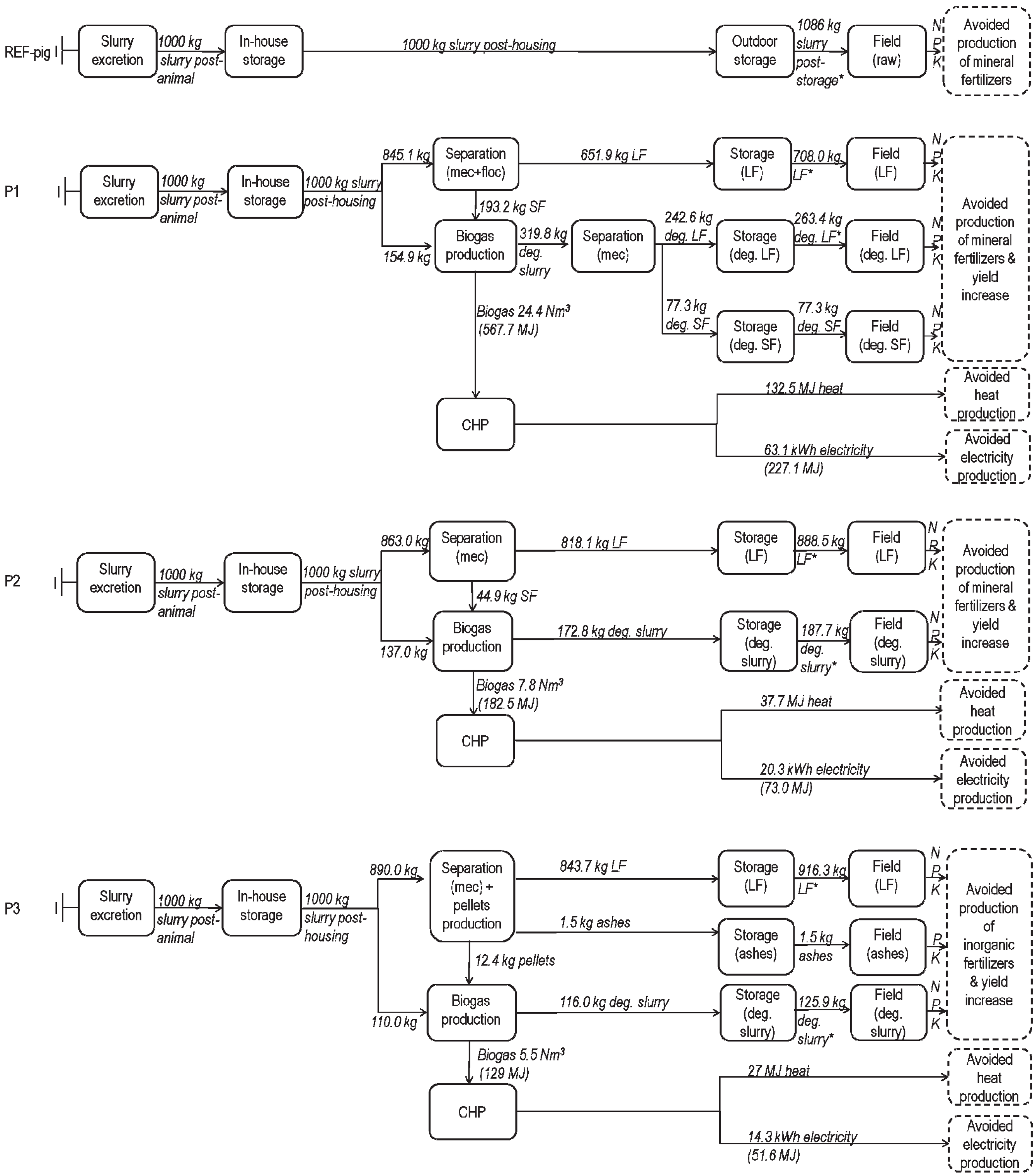

Figure 1. Process flow diagrams of the alternatives compared for pig slurry: (REF-pig) Reference system, (P1) Alternative P1: decanter centrifuge with polyacrylamide polymer, (P2) Alternative P2: screw press, (P3) Alternative P3: screw press and pellets fabrication. The dotted lines indicate avoided processes. Flows marked with * include the addition of rainwater. The diagrams are simplified and only include the main processes involved in the model. All flows are related to the functional unit. "Mec" stands for mechanical separation and "floc" for flocculants addition.

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

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, postanimal (kg ton^{-1} slurry postanimal)		6.60	1.1	2.9	77	37
REF-pig, posthousing (kg ton^{-1} slurry posthousing)[c]		5.48	1.1	2.9	70	33
REF-pig, poststorage (kg ton^{-1} slurry poststorage)		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 SI. [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 ref 18. [c] The REF-pig posthousing 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.

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 N m^3 CH_4 ton^{-1} VS. The solid fraction and fiber pellets obtained in alternatives P2 and P3, respectively, have a CH_4 yield of 187 N m^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 fiber 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 SI, 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 fiber 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 SI.

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 ref 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 SI.

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 modeling 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 poststorage in 2008.[16] The amount of mineral N avoided is thus 3.75 kg N ton^{-1} slurry poststorage 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 postanimal). This reflects how much less mineral N is applied per ton of slurry used as a fertilizer.

For alternative P3, it is slightly more, that is, 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 ref 30 and further explained in the SI.

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 SI. 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 (modeling details are presented in the SI). 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 toward 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.

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.

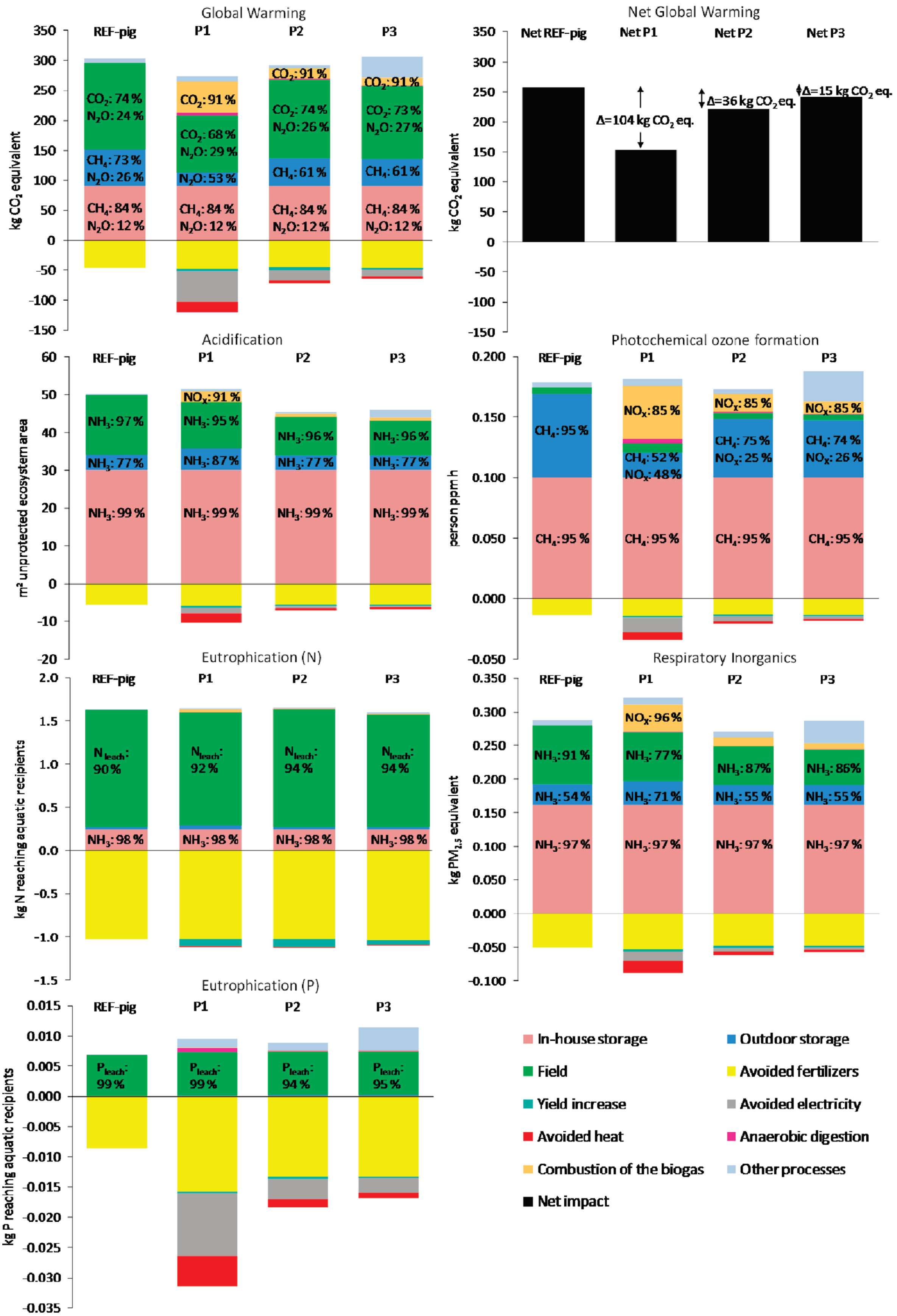

Figure 2. Breakdown of impact assessment results for all impacts and alternatives assessed.

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 postanimal)	31.71	19.45	27.62	25.41
C lost as CO_2 (field) (kg ton^{-1} slurry postanimal)	−30.68	−18.55	−26.98	−24.90
C stored in the soil (kg ton^{-1} slurry postanimal)	1.03	0.90	0.64	0.51
net CO_2−C "stored"[b] (kg ton^{-1} slurry postanimal)	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 postanimal 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., ref 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 bypassed 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 postanimal 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 SI. Ammonia emissions from in-house slurry storage has also been identified as an environmental hot spot in previous studies (e.g., ref 41) and mitigation measures to reduce NH_3 emissions from housing units have been the object of several studies (e.g., refs 42−44). Technologies allowing to reduce both CH_4 and NH_3 emissions from in-house slurry storage, for example, 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 ref 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, that is, 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 odors 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 evidence from several studies on cationic PAM, which are summarized in the SI, 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.

■ ASSOCIATED CONTENT

Supporting Information. Scope, functional unit, preconditions 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. This material is available free of charge via the Internet at http://pubs.acs.org.

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Paper V

Environmental Consequences of Different Carbon Alternatives for Increased Manure-Based Biogas

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Abstract

Manure-biogas is a renewable energy resource rather untapped in Europe in comparison to its full potential. Given the current and emerging renewable energy targets, considerable increases in its production can be expected. This consequential life cycle assessment (LCA) study investigated the environmental consequences of different co-substrate strategies for reaching drastic increases in manure-biogas production in Denmark. Six co-substrates not already fully used for biogas were considered: energy crops, straw, household food waste, commercial food waste, garden waste and the solid fraction deriving from source-segregation of animal urine and feces. Soil carbon changes as well as direct and indirect land use changes were included in the LCA. Source-segregated manure stood out as the environmentally best co-substrate, followed by garden waste. Co-substrates already in use for energy recovery (straw, household and commercial food wastes) displayed a more modest environmental performance while energy crops, here represented by maize silage, was the only option giving rise to net greenhouse gas emissions. This was essentially due to the indirect land use change emissions related to this scenario, which were quantified to 357 t CO_2 eq. ha^{-1} displaced.

Keywords

Anaerobic digestion; land use changes; food waste; garden waste; straw; life cycle assessment.

1. Introduction

Recovery of manure biogas is an acknowledged cost-effective mitigation technology for greenhouse gas (GHG) emissions in agriculture [1–3], being not only a source of renewable energy, but also a way to improve the GHG balance of traditional manure management systems. In the perspective of a fully renewable energy system, biogas also offers the possibility to be storable in the gas network, which provides flexibility for buffering the fluctuant energy supply from intermittent sources like wind and sun, as well as a fuel for transport.

In spite of that, the energy produced from manure-biogas in the European Union (EU) is far below its full potential, the ca. 50 PJ produced in 2007 from agricultural biogas plants (including other substrates than manure like energy crops and organic wastes) [4] representing less than 7% of the 827 PJ potential estimated for cattle and pig slurries alone [5]. A recent analysis of the national renewable energy action plans (NREAP) made by the European Member States in the framework of the renewable energy directive (RED) [6] nevertheless highlights that European Member States have provided ambitious biogas targets to meet their renewable energy obligations. In Denmark, for example, a target has been launched to achieve 50% use of manure for biogas by 2020 [7] as compared to the present use of only 5-7% [8].

Animal manures, however, are often too dilute with respect to their carbon (C) content, and it is a common practice for biogas plants to co-digest manures with C-rich substrates, in order to ensure a biogas production safeguarding the economic sustainability of the production [2,9]. On the other hand, using these co-substrates for boosting manure-biogas involves that these are taken away from their other applications, and the environmental consequences of this should be well understood in order to establish a sustainable strategy for achieving a colossal increase in manure-biogas.

This study aims to investigate the environmental implications of different C co-substrate alternatives for enriching manure biogas, using Denmark's target for a substantial increase in manure-biogas as a contextual framework. The substrates assessed were those considered to have the greatest potential to supply an increased manure-biogas production, namely: energy crops, straw, various biowaste types (household food waste, commercial food waste, garden waste), and the solid fraction deriving from source-segregation of animal urine and feces. Substrates already fully used for the manure being digested nowadays (e.g. industrial organic residues from fish, fruit, sugar, dairy or oil industries) were not considered.

2. Model Description and Key Parameters

2.1 LCA Model

The environmental impacts of the different co-substrate alternatives investigated were compared based on a consequential life cycle assessment (LCA) [10–13]. All input and output flows were related to a functional unit (FU) being the management of 1 tonne of freshly excreted pig manure (manure ex-animal). The slurry composition considered is presented in the supporting information (SI), as well as all emission data and mass balances related to the management of this raw slurry. The geographical scope considered for the LCA was Denmark, i.e. the data inventory for crop cultivation, manure management, and the applicable legislation were based on the Danish context. The life cycle impact assessment was carried out according to the Danish EDIP 2003 methodology [14] for the impact categories global warming (100 years horizon), acidification and aquatic eutrophication (distinguishing between nitrogen and phosphorus being the limiting nutrient for growth). Background (or generic) LCA data were based on the Ecoinvent v.2.2 database [15], and the assessment was facilitated with the LCA software SimaPro 7.3.3. Foreground (or system-specific) LCA data essentially included Danish-specific data for manure management (raw and digested), biogas production, crop cultivation and composition, co-substrates pre-treatment and energy conversion technologies, and are detailed in the SI.

2.2 System Boundary

Except for energy crops, all substrates considered in this study are waste products (i.e. manure, biowastes and straw) from other production systems. Based on the consequential LCA rationale, only processes that would react to a change in demand for manure-biogas should be included in the LCA. As any systems generating waste would, of course, be unaffected by the use of the waste, the processes upstream the generation of these wastes (e.g. the animal production system for manure, the crop production system for straw) were not included in the system boundary. For all scenarios, the system boundary thus starts with 1 tonne of raw manure as freshly excreted (ex-animal), which is afterwards temporally stored in-house before to be sent to a biogas plant (manure ab-housing). The CH_4 yield considered for manure ab-housing is 319 Nm^3 per t volatile solids (VS) [2]. Further, it was considered that co-substrates are added to this manure in order to get a mixture reaching a dry matter (DM) content of 10% after the first digestion step, and a carbon to nitrogen (C/N) ratio limited to 20, reflecting state-of-the-art practice of Danish biogas plants [2]. From the anaerobic digestion step, two outputs are produced: the digestate and the biogas. The biogas is assumed to be used for combined heat and power (CHP). In this study, the marginal electricity source displaced by the biogas was assumed to be from coal-fired power plants, and the marginal heat from natural gas based domestic boilers, based on [16]. The other output from the anaerobic digestion process, namely the digestate, was assumed to be stored in a concrete tank covered with a straw floating layer [2]. When appropriate, this digestate can be applied on agricultural fields as an organic fertilizer, thereby displacing mineral nitrogen (N), phosphorus (P) and potassium (K) fertilizers, considered to be calcium ammonium nitrate, diammonium phosphate and potassium chloride, respectively (marginal fertilizers). The modeling of fertilizer substitution is further detailed in the SI. Changes in soil C occurring as a result of applying the digestate on land instead of raw manure were estimated with the dynamic soil C model C-TOOL [17–19]. For all alternatives, the co-substrates (or the land required to cultivate it, in the case of energy crops), if not used for biogas, would have been used for other applications. Using the co-substrates for biogas thus divert them from their initial use, which implies a variety of consequences, among others that a substitute is needed to supply the service (e.g. energy, fertilizer) no longer provided by the co-substrates. In this study, this service no longer provided is referred to as the lost alternative, and the consequences of it (like the production of a substitute) are included in the system boundary. Similarly, it is considered that the raw manure used for biogas would have otherwise be conventionally stored and applied on land (reference slurry management), in the way described in [2]. The system boundary considered is illustrated in Figure 1, for the case of straw (scenario 2). System boundaries for all other scenarios are presented in the SI (Figures S1-S7).

2.3 Biogas Production

The biogas production considered in this study is based on a two-steps anaerobic digestion consisting of a completely stirred main digester and a post-digester from which ca. 10% additional CH_4 emissions are captured. It is assumed that the production is operated under mesophilic conditions, and that the biogas produced is constituted of 65% CH_4 and 35% CO_2, with a density of 1.158 kg Nm^{-3} biogas [2] and a LHV of 22.88 MJ Nm^{-3} biogas. Fugitive losses of 1% were assumed, based on recent LCA studies [2,20,21]. The biogas is considered to be burned in a biogas engine with efficiencies of 46% for heat and 40% for electricity [2], and it is assumed that only 90% of the net heat produced can substitute marginal heat, reflecting the losses occurring in periods with low heat demand [22]. Internal electricity consumption corresponding to 5% of the net electricity production [2,23] was assumed. Internal heat consumption was calculated considering that the mixture is heated from 8°C (Denmark's average annual temperature) to 37°C. The heat requirement was calculated considering a specific heat of 3.00 kJ kg^{-1} $°C^{-1}$ for the DM share of the input mixture, and of 4.20 kJ kg^{-1} $°C^{-1}$ for the water, based on [2].

3. Scenarios Description and Sensitivity Analysis

3.1 Scenario 1: Energy Crops

Maize silage has been chosen as the energy crop to represent this scenario given its high yield and its high C turnover efficiency [24]. It is considered to be produced in Denmark specifically for anaerobic digestion, and as such is displacing another crop [16], which is here considered to be maize for animal feed. Based on this, the additional hectares of maize needed for anaerobic co-digestion were modeled to displace hectares of maize used for feed. As the production of maize (for energy) instead of maize (for feed), which represents the direct land use change (DLUC) involved in this study, was assumed to result in negligible changes in emissions, the DLUC was excluded from the model (but considered in sensitivity analyses). Based on the consequential LCA logic, as well as on recent studies [25–27], the drop in supply of Danish feed maize resulting from this displacement will cause a relative increase in agricultural prices, which then provide incentives to increase the production elsewhere. Such increased crop production may stem from both increased yield and land conversion to cropland, the latter being also referred to as indirect land use change (ILUC) [25–27]. As illustrated in Figure S1 (SI), and as in recent ILUC studies [27–29], this study included the environmental impacts of the latter only. The life cycle inventory data used for maize were taken from [24] (for a sandy soil), which considered a yield of 12.05 t DM ha^{-1} y^{-1}. The maize composition considered is shown in the SI (Table S4). During storage of the maize silage, DM losses corresponding to 0.8% of the fresh weight are accounted for, based on the findings of [30]. When leaving the storage, the maize is considered to have a CH_4 yield of 382 Nm^3 per t VS, based on an average of Danish results [31,32]. ILUC was modeled as described in [16], and resulted in an emission of 357 t CO_2 eq. per ha feed maize displaced (Table S17), which was annualized over 20 y (i.e. to an annual figure of 18 t CO_2 eq. ha^{-1} displaced y^{-1}) in accordance with IPCC guidelines [33] as well as with the RED [34].

3.2 Scenario 2: Straw

Winter wheat straw is the most abundant straw in Denmark [35], and was therefore the straw type considered for this study. Its composition is presented in the SI (Table S21). Based on [36], a CH_4 maximum potential (i.e. at 100% degradation) of 432 Nm^3 CH_4 kg^{-1} VS was considered, as well as a biodegradability of 45%. Operations related to straw harvesting were modeled based on the life cycle inventory of [24], which considers a straw yield of 3.09 t DM ha^{-1}. During storage, losses corresponding to 1.1% of the initial DM were assumed, based on [37]. Given its considerable lignin content [36], straw is rather recalcitrant to microbial degradation. An extrusion pre-treatment (described in [38]) was therefore included in order to break the lignocellulosic structure of straw and render a maximum of its C content bioavailable. This was considered to increase straw biodegradability to 61% (i.e. 35% increase), and to consume 14.5 kWh per t of straw, based on the average results obtained by [38]. Straw combustion in a small-to-medium scale biomass CHP plants is assumed to be the lost alternative for this scenario. This type of straw management represents the fate of ca. 30% of the straw produced in Denmark [35], and this percentage is expected to increase given the renewable energy ambitions and obligations Denmark has [7,39]. Efficiencies of 27% and 63% for electricity and heat, respectively, are considered for straw combustion, based on [16]. The energy that is no longer produced because straw is now used for biogas would thus be produced by the fossil-based marginal heat and electricity considered, and this induced energy was modeled the same way as for the avoided energy due to biogas production (SI).

3.3 Scenario 3: Household Biowaste

This scenario involves the use of the food waste generated every year from the Danish households, also termed biowaste. The household food waste composition assumed is detailed in the SI (Table S58), and it is considered to have a CH_4 yield of 330 Nm^3 CH_4 t^{-1} VS, based on an average of Scandinavian studies [31,40–42]. Once collected, biowaste is separated from the overall household solid waste through a press separation technology, considered to recover 96% of the biowaste, and to require an electricity consumption of 10.6 kWh per t of biowaste output [43]. As required by EU legislation [44], food waste undergoes a hygienization process prior to biogas production. Under this process, the biowaste was considered to be heated from 8°C to 75°C, including a heat loss corresponding to 5% of the heat needed [45]. Based on [46], the hygienization process was assumed to have no effect on the CH_4 yield of the biowaste, nor on its composition. As more than 90% of the household biowaste generated in Denmark today is incinerated (SI), the lost alternative was considered to be the combustion of the biomass in a municipal solid waste incineration CHP plant, with electricity and heat efficiencies corresponding to 21% and 74% respectively, and a lower heating value (LHV) of 20 MJ kg^{-1} DM [41]. The emissions associated to this process are presented in the SI (Table S35). As for straw, the energy that is no longer produced because household biowaste is now used for biogas was modeled as marginal heat and power induced.

3.4 Scenario 4: Commercial Biowaste

This scenario considers commercial biowaste (or food waste) from wholesale and retail stores (SI). Based on the composition considered (Table S58), the methane yield of this biowaste was calculated to 277 Nm^3 t^{-1} VS. The life cycle processes and parameters considered for commercial biowaste are exactly the same as household biowaste.

3.5 Scenario 5: Garden Waste

Garden waste is generated during maintenance of public areas and private gardens. In this study, garden waste is assumed to be constituted of 75.6% easily degradable material (e.g. leaves and grass) and 19.5% branches, the rest consisting of woody parts, stones and foreign items, based on the findings of [47]. The full composition considered is detailed in the SI (Table S58). The methane yield assumed is 203 Nm^3 t^{-1} VS (Table S58). Before to be fed to the digester, garden waste is shredded, and a diesel consumption of 1.5 l t^{-1} waste was considered for this [48]. Open windrow composting was considered as the lost alternative for garden waste, being the fate of ca. 75% of the garden waste generated in Denmark (SI). The output of the composting process consists of screened wooden materials with 64% DM, and mature compost with 68% DM [49]. The compost is then stored in a completely covered storage facility and applied on land, while the wood chips are burned in a small-to-medium scale biomass CHP plant with the same efficiencies as for straw. The emissions considered for the composting process, the compost storage, the compost application on land and the wood chips burning are detailed in the SI. Induced mineral fertilizers due to the compost no longer spread, and induced marginal heat and power due to the wood chips no longer burned were considered in the model (SI).

3.6 Scenario 6: Source-segregated Manure

In this scenario, raw pig slurry is co-digested with the concentrated solid fraction obtained from source-segregation of urine and feces in the animal house (of a second farm). In this way, the substrate digested consists of manure only. The separation technology considered is based on the rotating belt conveyor technology developed by Lemay and coll. [50–52], which separates urine and feces immediately after excretion, preventing any significant contact between these, unlike most existing source-separation technologies [53,54]. The separation efficiencies and in-house ammonia (NH_3) emission reduction considered for this technology are presented in the SI. The separated solid and liquid fractions are stored temporarily in-house (less than 1 week), and the liquid is then stored in a covered storage tank (floating straw layer), while the solid is sent to a biogas plant. The emissions and mass balances taken into account for these processes are detailed in the SI. The methane yield considered for the solid fraction is the same as for raw manure, based on [2]. Just like the digestate, the stored liquid fraction is used as a fertilizer and applied on land, avoiding the production and use of marginal fertilizers. If not used for co-digestion, it is considered that the manure from the second farm would have never been separated, but merely managed according to the reference manure management. This is thus the lost alternative considered.

3.7 Scenario 7: Mono-digestion

A mono-digestion scenario was included for comparison purposes only. In this scenario, 1 t of raw pig slurry (ex-housing) is digested, which avoids the reference slurry management to occur. Processes and parameters for this scenario are as previously described for the raw manure portion of each scenario.

3.8 Sensitivity Analysis

Two main types of sensitivity analysis were performed. The first one consisted to consider different lost alternatives for selected scenarios, namely: straw plowing (instead of combustion) for the straw scenario, landfilling (instead of combustion or composting) for the three biowaste scenarios, and mono-digestion (instead of conventional manure management) for the source-segregation scenario (i.e. assuming that if not separated, the manure from the second farm would have been mono-digested). The second type of sensitivity analysis consisted to test different variants for the maize and straw scenarios. In the case of maize, it consisted to assume that i) spring barley and ii) sugar beet were displaced instead of feed maize (i.e. different DLUC were considered). One additional variant for maize was to consider a natural on-field drying prior to harvest, so its DM content could rise from 31% (baseline case) to 40%, thereby allowing to reach a mixture manure-maize with 10% DM (as opposed to 6.6% in the baseline scenario). For straw, two different pre-treatments were considered, namely an alkali pre-treatment as well as a pre-treatment combining straw explosion and enzymatic hydrolysis (instead of the baseline extrusion pre-treatment). The modeling considerations behind these sensitivity analyses are presented in the SI.

4. Results and Discussion

4.1 Biogas Production

Each scenario required different amount of co-substrates per tonne of fresh manure excreted (FU), and consequently, different quantities of biogas were produced for each of them (Table 1). As shown in Table 1, scenarios requiring the lowest share of manure in the input mixture (maize and commercial biowaste) were also those with the greatest amount of biogas produced, and consequently those with the greatest benefits from substituting marginal energy (gray and red bars in Figure 2).

Table 1. Co-substrates required, biogas produced and input mixture characteristics for each alternative[a].

	Maize	Straw	HW	CW	GW	SSM	Mono
Co-substrates required per FU (kg ww)[b]	1303	183	785	1701	297	1441	-
Biogas produced per FU (Nm^3)	250	87	131	193	75	194	27
C/N ratio of input mixture	19.7	17.1	11.9	13.2	13.1	10.1	6.5
DM of mixture after 1st digestion step (%)	6.6	10.0	10.0	10.0	10.0	10.0	3.7
Share of manure in input mixture (%)	43	85	56	37	77	100[c]	100
Nm^3 CH_4 produced per t mixture	70	48	48	46	38	52	18

[a] HW: household food waste; CW: commercial food waste; GW: garden waste; SSM: source-segregated manure; Mono: mono-digestion.
[b] This consists of the silage maize as leaving the storage, the straw as leaving the extrusion pre-treatment, the household and commercial biowaste as leaving the hygienization process, the garden waste as leaving the shredding process and the source-segregated manure as leaving the in-house storage. ww: wet weight
[c] Consisting of 41% manure ex-housing and 59% source-segregated solid fraction.

4.2 LCA Results

Figure 2 highlights that for all impact categories, using source-segregated manure as a co-substrate for biogas was the scenario with the lowest environmental impacts. This is mainly due to the lost alternative of this co-substrate, which would have otherwise been managed as raw manure. It thus emphasizes the tremendous environmental benefits of mitigating the emissions of the traditional manure management (here particularly storage and spreading), as also highlighted by previous studies [2,23].

Global warming results (Figure 2a) highlight garden waste as a co-substrate strategy allowing a significant reduction of GHG. Once again, this is partly explained by the lost alternative of this co-substrate, which is composting. In this case, composting the 297 kg of garden waste needed for biogas (Table 1) would have resulted in 157 kg CO_2 eq. per FU (which includes the credit for the recovered energy from the incinerated wood chips portion of the waste, Table S72 and Figure S13), highlighting it is better to prioritize it, GHG-wise, for manure-biogas. This conclusion was also drawn by [55] for a similar waste stream (roadside grass).

Figure 2 shows that the maize scenario is the only one giving rise to a net global warming impact. This result is mostly due to the ILUC it involved ("co-substrate lost alternative" bar), which raises the discussion around the accuracy of both the land expansion figure considered, i.e. the net area expanded per ha of feed maize displaced and the GHG figure considered, i.e. the net CO_2 emission due to arable land expansion. For the former, this study considered 2.35 ha transformed per ha of feed maize displaced in Denmark, corresponding to 13.9 ha transformed per TJ of energy in the biogas produced (SI). Similar figures were presented in others studies, e.g. 10.7 ha expanded per TJ in [27] (average for different biofuel systems) or 10.8 ha expanded per TJ in [56] (soy biodiesel), although other studies (e.g. on corn ethanol systems) presented rather smaller figures (e.g. 3.5 ha per TJ in [57] or 1.5 ha per TJ in [28]). It should be noted that the expansion figure considered in this study would have been lower if e.g. barley would have been the displaced crop (sensitivity analysis section), reflecting that the greater the yield of the crop displaced is, the greater the amount of carbohydrates no longer available on the feed market is, so the greater is the land expansion needed to compensate this lack. Compared to other publications where ILUC GHG figures per hectare displaced are presented, the figure used in this study, i.e. 18 t CO_2 eq. ha^{-1} displaced y^{-1}, appears in the middle-high range. For example, [29] reported a range of 10-28 t CO_2 eq. ha^{-1} displaced y^{-1} in relation to the implementation of the RED in Europe [34]. On the other hand, [23] reported figures of 2.6-10 t CO_2 eq. ha^{-1} y^{-1} for turning hectares of German grassland to cropland production. However, it is rather unlikely that the overall ILUC response of a strategy requiring more maize for manure-biogas would result in temperate grassland only being converted to agriculture. In fact, as highlighted in [26,58–61], biomes like tropical or temperate forests containing 50-89% more C than temperate grassland [25] are those that are likely to be the first converted following an increase demand for crops, these being the biomes where the frontier between agriculture and nature is already moving. Further, it should be highlighted that the ILUC figure used in this study does not reflect the GHG related to the intensification of crop production, which accounts, based on the results of [26], to ca. 30% of the displacement response. Emissions from intensification, particularly if the intensification response occur through additional N fertilizers inputs, may in fact be considerable [27,62], and even greater than the emissions from land expansion [61]. To summarize, it is acknowledged that the overall quantification of the ILUC impact remains an uncertain figure. However, as stressed by [63], it is not zero, and as such must be estimated based on best of available knowledge whenever energy crops are involved.

The straw and biowaste scenarios all allowed a net GWP reduction, which highlights that GHG-wise, these co-substrates are better used in biogas than in their previous use (i.e. combustion or composting). It should also be highlighted that co-digestion with manure has two additional advantages over incineration: i) it allows to recycle these co-substrate's nutrients, including the slowly degradable C, which are essentially lost in the incineration case, and ii) it produces a storable gas, a key flexibility asset for a renewable energy system. Further, using these co-substrates for increased manure-biogas allows to minimize (or prevent) the amount of energy crops used as co-substrates, and thereby the indirect land use changes related to it.

Figure 2 also points out digestate/liquid fraction handling as a "hot spot" for all impact categories. For global warming, the digestate/liquid fraction' impact is essentially due to biogenic CO_2 (57-66%), reflecting the emission of the biogenic C back to atmosphere. In fact, based on the soil C model used as well as on the grain-rich rotation assumed (SI), between 71-100% of the C in the organic fertilizers involved in this study ended up to be emitted to the atmosphere (the rest entering the soil C pool), over a time horizon of 20 y. For the acidification impact category (Figure 2b), the impact of the digestate/liquid fraction is essentially due to ammonia (NH_3) (over 98% for all scenarios). One way to significantly reduce NH_3 releases from the digestate/liquid fraction handling would consist to acidify the digestate/liquid fraction, a well-known technology [64–66] that allows keeping inorganic N in the ammonium form instead of the volatile NH_3 form, which would cancel off the differences between most scenarios for the acidification impact category.

Results for aquatic eutrophication (N and P) reflect that the digestates from the maize, household biowaste, commercial biowaste and source-segregated manure scenarios are those involving the highest amounts of N and P applied per FU (Table S65). For source-segregated manure, this is however cancelled out because of the lost alternative, consisting to spread raw manure (Figure 2f). For N, the model already considers that N is applied up to the strict crop requirements, based on the Danish regulations for fertilization [67,68], but nitrate losses could nevertheless be minimized e.g. through the addition of catch crops in the crop rotation [24]. For P, the losses could be reduced through precision dosing, i.e. applying the digestates with high P content (per tonne digestate; Table S64) in areas with P deficits. Overall, the maize and commercial biowaste scenarios were those resulting with the highest eutrophication potentials (for both N and P).

The results of this study are expressed per tonne of excreted manure, being the FU selected. To be comparable with other studies, results were also expressed, for the global warming impact, per Nm^3 biogas produced and per tonne of DM input to the digester (Figure 2). Figure 2 illustrates that the results present the same overall ranking independently of the functional unit, which confirm the reliability of the results presented per FU. Results per Nm^3 biogas produced highlighted the meaningfulness of avoiding conventional manure management, but failed to reflect that some scenarios allowed the production of more energy than others. Results expressed per tonne DM input did not provide much additional insights, essentially reflecting the differences in the proportion of DM supplied by manure in each scenario.

4.3 Sensitivity Analysis Results

As illustrated in Figure 3a, the lost alternative has significant importance on the absolute results. If straw would have otherwise been plowed down, the straw scenario would then present more savings than garden waste, GHG-wise. Avoiding landfilling instead of incineration would be an advantage for household but not for commercial biowaste, which highlights the poor environmental performance of commercial biowaste incineration, a waste with a rather low DM content. This is further detailed in the SI (Figure S13 and Table S75). For garden waste, Figure 3a highlights the GHG advantage of avoiding composting, though both incineration and landfilling as lost alternatives would have also led to GHG savings superior to those of mono-digestion. If the source-segregated manure would have otherwise been digested (mono-digestion), as this would be the case in a future where all excreted manures would undergo anaerobic digestion, savings (in terms of global warming) would still be obtained, although in much smaller magnitude.

DLUC was not included in the baseline scenario since it was assumed that the differences between cultivating maize (for energy) versus maize (for feed) were insignificant. Figure 3b illustrates, through the “DLUC” bar, the differences between cultivating maize and cultivating barley or sugar beet. In both cases, this reflects the differences in yield for these crops. For sugar beet, whose yield is ca. 3% greater than maize (Table S71), the net DLUC is positive, which increases the overall GWP of this scenario. This essentially reflects that the overall GHG emissions of the cultivation system are smaller for sugar beet than maize, as shown in [24]. For barley, whose yield is ca. 180% lower than maize (Table S71), the main effect is however the ILUC effect, i.e. it reflects that as less crops (and thus carbohydrates) are then displaced, less land conversion is needed to replace the carbohydrates no longer supplied from Denmark. However, in the perspective of a high bioenergy future like the one modeled in this study, it is likely that there will quickly be no more low-yielding crop like barley to offset, in which case this DLUC benefit could not be applied.

As opposed to all other baseline scenarios, the maize scenario could not reach the 10% DM input (Table 1), as a result of the high water content of maize silage. Revoking this limit by considering a natural drying of the maize to 40% DM and by adding N in the mixture to ensure a C/N limited to 20 however worsened the net GWP of this scenario. This scenario would lead to a biomass mixture input consisting of 74% maize and 26% manure. Though this increased the energy production, it also increased significantly the ILUC, as well as the GHG from the digestate handling (Table S76). On a global warming perspective, this solution is therefore of little interest, even if more energy is produced. Finally, as highlighted in Figure 3b, there are no clear differences, GHG-wise, between the different straw pre-treatments. For the combined straw explosion and enzymatic hydrolysis pre-treatment, it should however be mentioned that the obtained results are conditional to a DM input of 6.8% (which was needed for ensuring a C/N ratio of maximum 20). A greater share of such pre-treated straw would involve greater energy input for the pre-treatment, among other considerations.

4.4 Limits

Nutrients' recycling through land application of the digestates was considered for all scenarios, assuming all digestates met the legal quality requirements in force in Denmark for land application [67–69]. In practice, this may not be the case for all digestates, which especially applies for the biowaste scenarios. In fact, these were assumed to be separated through a mechanical separation process (press), which may, in practice, induce contamination (e.g. heavy metals, visible contaminants like plastics and metals) that would render the digestate unsuitable for land application. If biowastes are considered as part of the strategy for the Danish manure-biogas ambitions, digestate' analyses should therefore be performed to verify its compliance with norms for application on land, and this in the light of various separation techniques for getting the biowaste. Another limit of the study consisted of the quality of the data used, as quality data (e.g. composition, CH_4 potential, LHV) was rather limited for some substrates, especially commercial biowaste. At the light of a recent study [70], it also appears that the methane yield considered for garden waste was rather conservative, which implies that the environmental benefits of this scenario may be even greater than estimated in this study. Further, the study could also be broadened through investigating additional slurry types (e.g. cattle, mink), additional co-substrates (e.g. deep-litter, algae, natural grasslands) as well as additional uses of the biogas (e.g. transport, injection in the gas grid). Moreover, additional strategies for a drastic increase in manure-based biogas could be assessed, like for example through the use of recycled carbon dioxide from stationary combustion processes and hydrogen from the electrolysis of water, powered by surplus electricity (e.g. wind).

5. Conclusion

The life cycle assessment performed in this study assessed the environmental consequences of different C co-substrates alternatives for enhancing the CH_4 (and thus the energy) produced from manure-based biogas. It highlighted the tremendous environmental benefits, particularly with respect to global warming, of using manure for biogas, instead of managing it "conventionally". As a result, important additional benefits were obtained for the co-substrates scenarios allowing to use more manure for co-digestion. For this reason, source-segregated solid manure (i.e. obtained from preventing any contact between animal urine and feces) was highlighted as the co-substrate yielding the greatest environmental benefits overall, and as such appeared as the most sustainable strategy for a drastic increased manure-biogas production. Maize silage was the co-substrate scenario allowing to produce the greatest amount of biogas per functional unit. However, it also was the only option giving rise to a net increase of greenhouse gas emissions, because of the ILUC it involved. Straw and biowastes (i.e. garden waste as well as household and commercial food waste) all allowed a net GWP reduction, reflecting that GHG-wise, these co-substrates are better used in biogas than in their previous use (i.e. composting or combustion with energy recovery). In the perspective of a drastic increase of manure-based biogas, source-segregated manure, straw and biowaste should thus be prioritized as co-substrates for anaerobic digestion. On the other hand, relying on a long-term strategy involving energy crops would, based on the results obtained in this study, end up in more environmental impacts (global warming, particularly) than the fossil reference.

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Figures

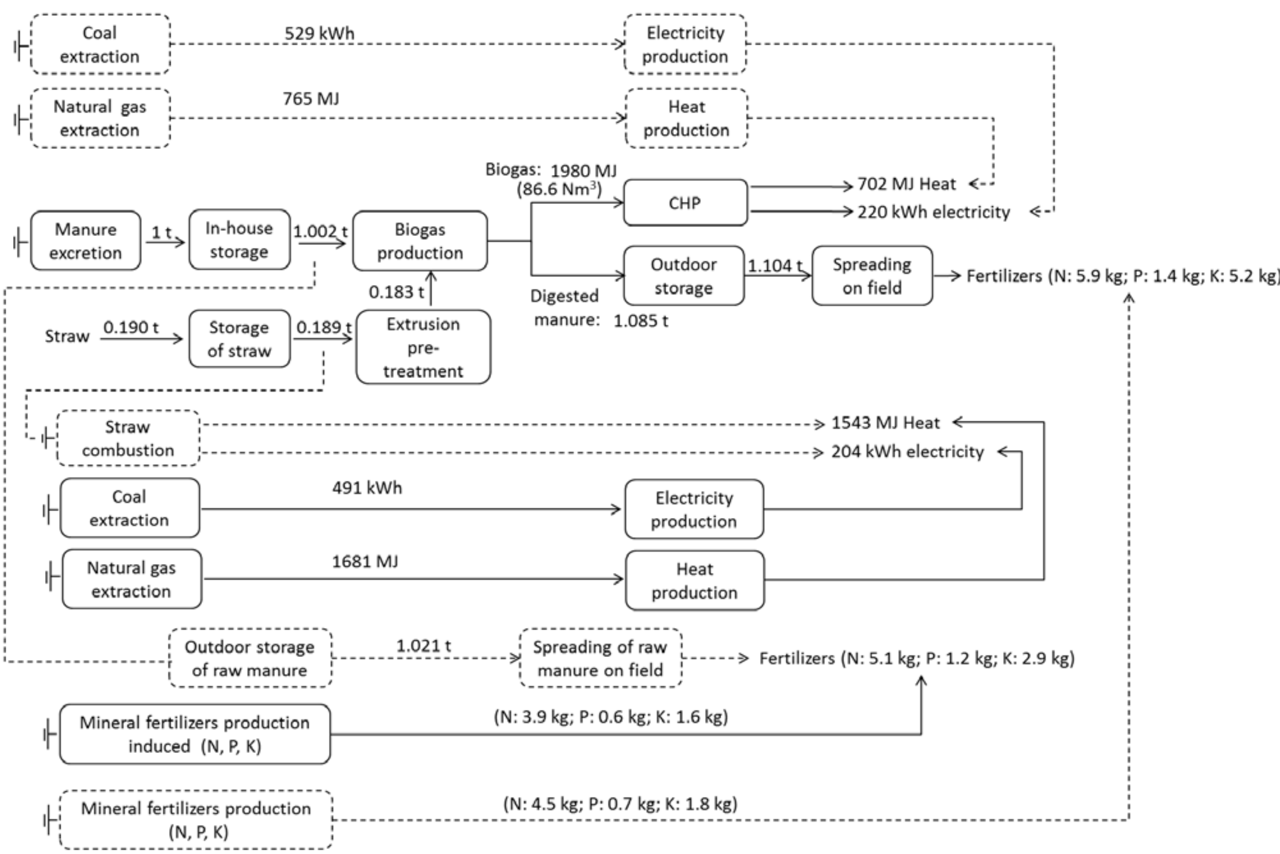

Figure 1. Process flow diagram for scenario 2 (straw). Dotted lines indicate avoided flows. All flows are related to the functional unit, i.e. 1 tonne of manure excreted.

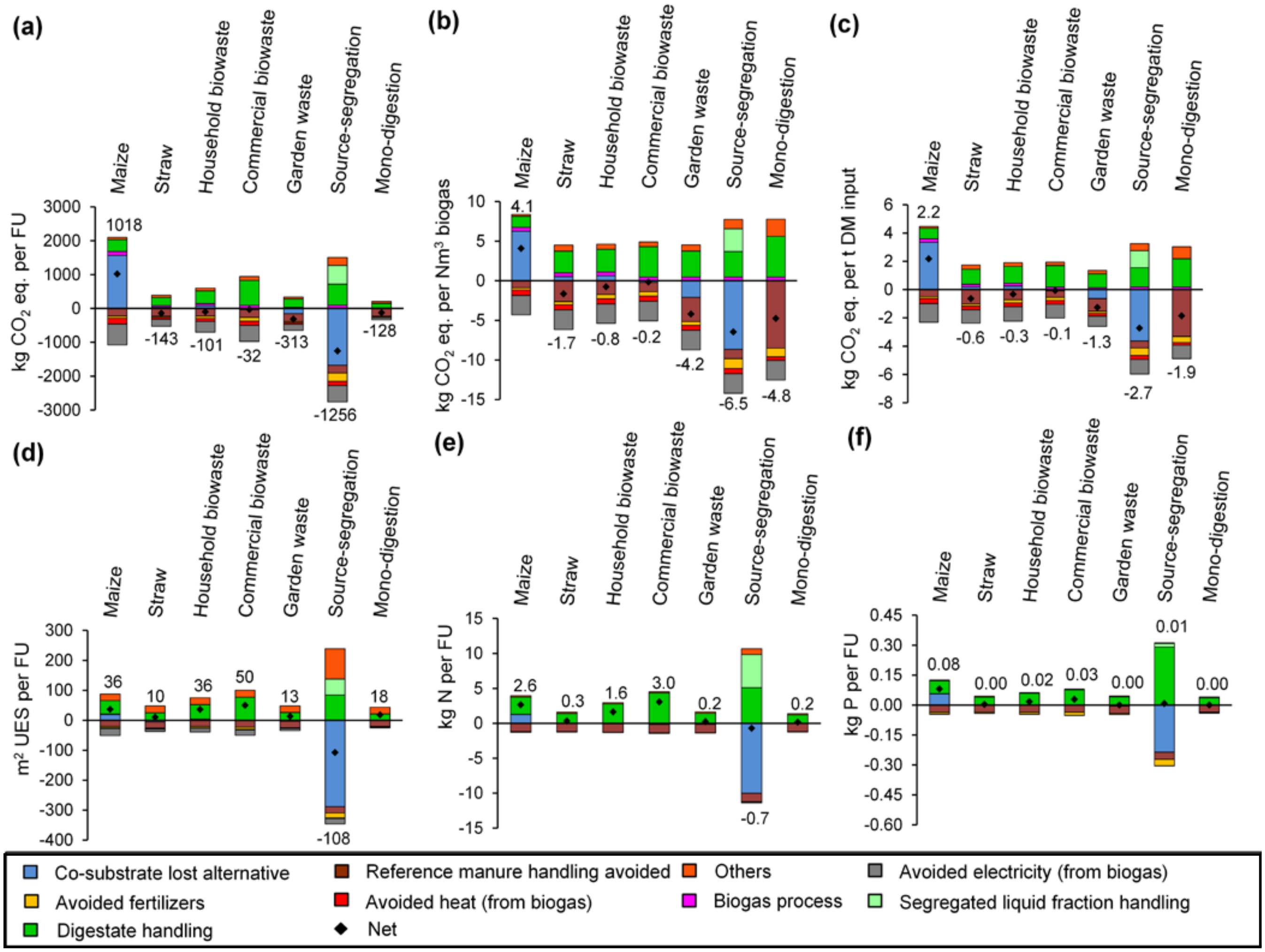

Figure 2. LCA results for the baseline scenarios a) global warming, per FU; b) global warming, per Nm^3 biogas; c) global warming, per tonne DM input; d) acidification, per FU (m^2 unprotected ecosystems eq.); e) aquatic N-eutrophication, per FU; and f) aquatic P-eutrophication, per FU. Global warming results are for a 100 y horizon.

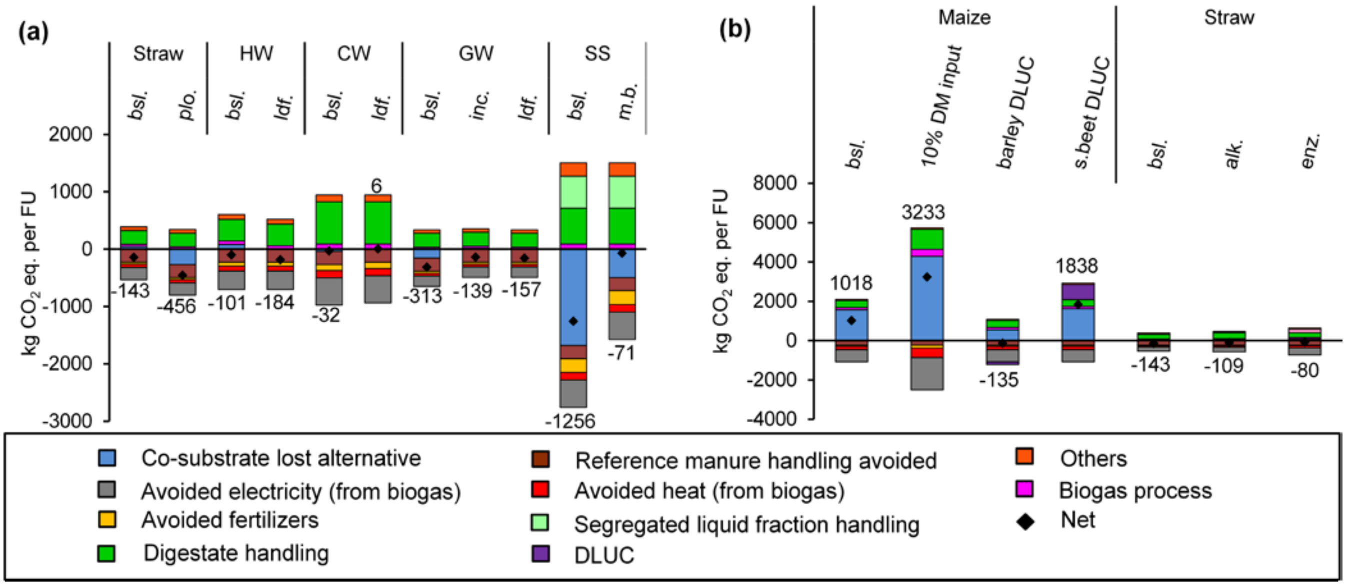

Figure 3. LCA results for the global warming impact, for the sensitivity analyses performed a) with different "lost alternatives" for selected scenarios; b) with different variants for the maize and the straw scenarios. Where "bsl" is the baseline scenario; "plo" is straw ploughing; "ldf" is landfilling; "inc" is incineration; "m.b." is mono-digestion biogas; "alk" is the alkali pre-treatment and "enz" is the combined straw explosion-enzymatic hydrolysis pre-treatment. HW: household biowaste; CW: commercial biowaste; GW: garden waste; SS: source-segregated manure.

Appendixes

APPENDIXES

This PhD thesis comprises 4 appendixes:

- Appendix A: Supporting Information to Paper ***I***, as available online
- Appendix B: Supporting Information to Paper ***II***, as available online
- Appendix C: Supporting Information to Paper ***IV***, as available online
- Appendix D: Supporting Information to Paper ***V***, (*submitted*).

These appendixes consist of the background documentation behind papers ***I***, ***II***, ***IV*** and ***V***. It includes, among others, most mass balances performed as well most calculation details.

The appendixes are not included herein, but can be accessed free of charges online:

- Appendix A: http://onlinelibrary.wiley.com/doi/10.1111/j.1757-1707.2012.01174.x/suppinfo
- Appendix B : http://pubs.acs.org/doi/suppl/10.1021/es3024435
- Appendix C : http://pubs.acs.org/doi/suppl/10.1021/es200273j

Appendix D is not yet available online, but is available upon request.