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Published in:
Science of the Total Environment

Link to article, DOI:
10.1016/j.scitotenv.2017.01.207

Publication date:
2017

Document Version
Peer reviewed version

Citation (APA):
Parajuli, R., Knudsen, M. T., Djomo, S. N., Corona, A., Birkved, M., & Dalgaard, T. (2017). Environmental life cycle assessment of producing willow, alfalfa and straw from spring barley as feedstocks for bioenergy or biorefinery systems. *Science of the Total Environment*, *586*, 226-240. https://doi.org/10.1016/j.scitotenv.2017.01.207

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Environmental Life Cycle Assessment of producing willow, alfalfa and straw from spring barley as feedstocks for bioenergy or biorefinery systems

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Abstract:

The current study aimed at evaluating potential environmental impacts for the production of willow, alfalfa and straw from spring barley as feedstocks for bioenergy or biorefinery systems. A method of Life Cycle Assessment was used to evaluate based on the following impact categories: Global Warming Potential (GWP_{100}), Eutrophication Potential (EP), Non-Renewable Energy (NRE) use, Agricultural Land Occupation (ALO), Potential Freshwater Ecotoxicity (PFWTox) and Soil quality. With regard to the methods, soil organic carbon (SOC) change related to the land occupation was calculated based on the net carbon input to the soil. Freshwater ecotoxicity was calculated using the comparative toxicity units of the active ingredients and their average emission distribution fractions to air and freshwater. Soil quality was based on the change in the SOC stock during the land use transformation (from Danish forestry) to an arable land. Environmental impacts for straw were economically allocated from the impacts obtained for spring barley. The results obtained per ton dry matter showed a lower carbon footprint for willow and alfalfa compared to straw. It was due to higher soil carbon sequestration and lower N_2O emissions. Likewise, willow and alfalfa had lower EP than straw. Straw had lowest NRE use compared to other biomasses. PFWTox was lower in willow and alfalfa compared to straw. A critical negative effect on soil quality was found with the spring barley production and hence for straw. Based on the energy output to input ratio, willow performed better than other biomasses. On the basis of carbohydrate content of straw, the equivalent dry matter of alfalfa and willow would be higher. The environmental impacts of the selected biomasses in biorefinery therefore would differ based on the conversion efficiency, e.g. of the carbohydrates in the related biorefinery processes.

Keywords: Energy crops, biorefinery feedstock, land use change, toxicity, environmental sustainability, biomass utilization efficiency

1. Introduction:

Increasing demands for food, feed, fibers and energy from the available agricultural land has stressed to optimize the biomass productions from the available land. It has also stressed to explore sustainable opportunities for the combined production of fuels, food/feed and chemicals (Parajuli *et al.*, 2015a). Biorefineries thus evolved to bring new value chains in the biomass conversion by producing cascades of biobased products. The types of biomass used is additionally important for their sustainable conversion to biofuels (Caputo *et al.*, 2005), since their different chemical composition (e.g., carbohydrate content) affecting the biochemical conversions (Stephen *et al.*, 2012). One of the crucial challenges for sustainable biorefinery operation is maintaining a year-round supply of biomass (Cherubini *et al.*, 2007). This is relevant as over exploitation of biomasses would be on: soil carbon (C) sequestration (Fargione *et al.*, 2008), nitrous-oxide emissions (Crutzen *et al.*, 2008), nitrate pollution (Donner and Kucharik, 2008), biodiversity (Landis *et al.*, 2008) and human health (Hill *et al.*, 2009). Likewise, soil quality is crucial for the long-term productivity of agricultural soil and also for the provision of other ecosystem services (Milà I Canals *et al.*, 2007a). Soil quality is often assessed in terms of soil organic carbon change and fertility (Lal, 2015). Likewise, sustainable management of available resources is also pertinent. Estimates show that about 10–20% of existing grassland within the EU member states, approximately 16.4 million hectare (Mha), is available for alternative uses to animal feed production (Mandl, 2010). These have stressed to diversify the supply of biomass to different biorefinery systems so that sustainable production of both fuel and non-fuel products is possible.

Life Cycle Assessment (LCA) has been widely used as a tool for assessing the environmental sustainability of different production systems (European Commission, 2015). Most of the LCA studies related to biomass production system have mainly focused on greenhouse gas (GHG) balances. In order to select the right biomasses and processing methods, it is also necessary to evaluate other impact categories besides GHG and energy balances (Wagner and Lewandowski, 2016). These are helpful to avoid creating flawed decision support tools for biorefining policies that may occur if evaluations are based on a single indicator (Finkbeiner, 2009). In most of the LCA studies, combinations of different crops including annual and perennial grasses were partially covered and described. Mogensen *et al.* (2014) quantified the impacts of producing different crops for livestock production, but mainly focussed on the carbon footprint. Likewise, Pugesgaard *et al.* (2013) compared the energy balance and nitrate leaching of annual crops and grasses in a rotation. Impacts of Soil Organic Carbon (SOC) on the GHG balance was also partially addressed in most of the identified studies (Tonini *et al.*, 2012). In a study of Short Rotation Coppice (SRC), Dillen *et al.* (2013) focused on energy balance, but assumed a less intensified farming system. Similar studies on SRC include Goglio and Owende (2009), Pugesgaard *et al.* (2015) and Sabbatini *et al.* (2015), but they were based on different assumptions with regard to farming system. Gallego *et al.* (2011) was limited for not covering SOC change in the overall GHG balances of alfalfa production. Godard *et al.* (2013) compared six feedstock supply scenarios, but the emission factors and other basic assumptions adopted in their modeling were less consistent with our study, particularly regarding system boundary and the agro-climatic conditions. Wagner and Lewandowski (2016) included a wide range of impact categories in their study, but it seemed that the system boundary for the related emissions was differently used, e.g. for the calculation of freshwater ecotoxicity. Birkved and Hauschild (2006) suggested that emissions of pesticides to soil can occur indirectly, hence it is relevant to assess the relative emissions to air and freshwater. Parajuli *et al.* (2016) using the tool PestLCI 2.0.6 presented the sensitivity of using different types of pesticides and varying agro-climatic parameters on the emission distribution fractions of the active ingredients. Based on their study, ecotoxicological measures were sensitive to the types of active ingredients and the season of applying the pesticides, as also coined in similar line in Dijkman *et al.* (2012).

Environmental sustainability assessments of the biomass production is one of the first steps to be taken for ensuring sustainable diversification in their supplies and the conversions (Parajuli et al., 2015a). In this study, LCA is used for evaluating the environmental footprints of producing willow, alfalfa and straw from spring barley. The biomasses were selected on the basis of their different physio-chemical and environmental qualities (Parajuli *et al.*, 2015b). Higher cellulose to lignin ratio in straw and willow can be regarded as a quality that qualifies them for sugar-based biorefinery platforms. Likewise, the crude protein and carbohydrate contents of alfalfa make it suitable for a green biorefinery technology to produce green protein and other biochemicals (e.g. lysine, lactic acid) (Parajuli et al., 2015b). Straw is regarded to induce a lower land use competition compared to other feedstocks (Kim and Dale, 2004). Willow, in turn, is suited for cultivation on marginal land, reducing its competition with food crops grown on fertile land (Helby *et al.*, 2004). Willow also has an effective nutrient uptake from soil, lower GHG emission and better fossil fuel energy balance compared to fossil fuels (Murphy *et al.*, 2014). The current study hence aims at evaluating different types of biomass feedstocks taking into account the important environmental impact categories.

2. Materials and Methods:

2.1. Goal, system boundaries and functional unit

The primary goal of this study is to provide a holistic view of resource requirements, emissions and finally evaluating environmental impacts for the production of the selected biomasses for utilizing them as bioenergy or biorefinery feedstocks. For this purpose, we take into account the system-wide effects of resource utilization starting from material extraction, processing, production and their utilization in an agricultural system. The system boundaries for the production of the selected biomasses are shown in Figure 1. The system boundaries covered: (i) the background system (upstream processes) and (ii) the foreground system (downstream processes). The background system included the production of the assumed material inputs (e.g. fuel, chemicals, and agricultural machinery) and their supply to the foreground system. All the necessary data related to the background system were based on Ecoinvent 3 (Weidema *et al.*, 2013), unless otherwise stated in the text below. Foreground system included the actual farm operation activities and the related emissions during the production of the selected crops. Data for the foreground system are elaborated in the section 2.3.

The functional unit (FU) of the assessment is 1 tonne dry matter (t DM) of the harvested biomasses. Storage of the biomasses is not accounted within the system boundary. The results of the environmental impacts are also shown in terms of energy in gigajoule (GJ) of the harvested biomasses.

Figure 1: System boundaries for the selected biomasses and related elementary flows. (Figure 1a represents the general system boundary and Figure 1b represents the production cycle of willow.)

2.2. Environmental impact categories and the assessment methods

The environmental impact categories with their units are: (i) Global Warming Potential in 100 years (GWP_{100}) (kg CO_2 eq), (ii) Eutrophication Potential (EP) (kg PO_4 eq), (iii) Non-Renewable Energy (NRE) use (MJ eq), (iv) Agricultural Land Occupation (ALO) (m^2), (v) Potential Freshwater Ecotoxicity (PFWTox), expressed as ‘comparative eco-toxic units’ (CTU_e) and (vi) Soil Quality (t C). These potential impacts were evaluated with respect to the FU of the study.

The “EPD” method (Environdec, 2013) was used for the assessment of the first three impact categories, while ALO was assessed using the ReCiPe method (Goedkoop *et al.*, 2009). PFWTox was calculated using the ILCD method (European Commission, 2012), and emission distribution fractions of the pesticides at the farm level were based on the study reported by Parajuli et al. (2016). The choice of different impact assessment methods was mainly due to following two reasons: (i) to cover most of the selected impact categories by single method and (ii) to interpret the results of the life cycle impact assessment, in the expressed units of the selected impact categories, as described above. For the former point the EPD fulfilled by covering the first there impact categories. The difference in the impact assessed, e.g. GWP_{100} or climate change in EPD and ILCD respectively was nominal. Likewise, the ILCD method has implemented all the USEtox factors (Rosenbaum *et al.*, 2008) that are suggested for calculating the ecotoxicological measures (European Commission, 2012). The method also interprets the result in terms of CTU_e, as in the USEtox model. This offers flexibility to the researchers to use either of the methods and interpreting the results on the basis of common unit. Moreover, ISO (2006) also does not recommend a specific method, suggesting that the choice should be based on the specific requirements of the user (European Commission, 2010).

With regard to soil quality, it was considered as an environmental impact category, in accordance to Brandão *et al.* (2011). SOC stock change (Δ SOC stock) was used as an indicator of soil quality (IPCC, 2000; Milà I Canals et al., 2007a). The impact was defined as a carbon deficit (or credit, indicated by negative values) with the unit 't C·year', giving the amount of extra carbon temporarily added to or removed from the soil compared to a reference system of a study (Milà i Canals *et al.*, 2007b).

2.3. Life Cycle Inventory Analysis

2.3.1. Crop production data

Table 1 shows the detailed Life Cycle Inventory (LCI) for the production of the selected biomasses. All the material inputs (agro-chemicals, fuel, energy, etc.) were estimated on an annual basis. These inputs were calculated from the total inputs estimated during the crop production life cycles and were divided by their respective number of life cycle years.

With regard to straw production, the material inputs and the environmental burdens were economically allocated from the production of spring barley. The allocation factor was 19% to straw, calculated based on sale prices for straw and cereals for the period 2011-2015 (SEGES, 2015). The quantity of seeds for producing spring barley was based on Jørgensen *et al.* (2011). Yield of straw was 55% of the grain yield (Taghizadeh-Toosi *et al.*, 2014a). The grain yield was based on average figures of spring barley cultivated on Danish sandy soil (Oksen, 2012; Statistics Denmark, 2013). The frequencies of farm operations (tillage, application of agro-chemicals and harvest) were all based on Jørgensen et al. (2011), or otherwise stated in the text below. The application of synthetic fertilizer (N, P, K) followed the Danish regulations (NaturErhvervstyrelsen, 2015). The amount and type of pesticides, i.e. the active ingredients (a.is), assumed for spring barley were based on the actual practice on Danish farms, as summarized in Ørum and Samsøe-Petersen (2014). Details on the application of the selected pesticides over the crop production life cycle years are given in Table S.4 of the Supporting Information (SI).

Production of willow was divided into two stages: (i) production of cuttings, (ii) production of the main crop. The main crop production included the farm operations: field preparation (tillage and application of agro-chemicals), planting of cuttings, harvesting and field restoration at the end of the life cycle of 22 years (i.e. also including the cuttings production) (Figure 1.b). The planting density was set to 12,000 cuttings ha^{-1} (Sevel *et al.*, 2012) and material inputs for the cutting production are shown in SI3 (Table S.3). After the cutback process the cuttings were transported for the plantation to a distance of 3 km (to the farm, single trip). The weight of the cuttings was 20 g cutting^{-1} (Rewald *et al.*, 2016). The annual application rate of pesticides for the production of willow was calculated from its total recommended life cycle dose (SEGES, 2010) (see SI, Table S.4). The first fertilizer application was assumed to take place after field preparation, since it has a tendency to lower the potential nitrate leaching (Heller *et al.*, 2003). Fertilization after planting was assumed to be carried out in every harvest-year and a year after each of the harvest-years. This amounted to 13 applications per ha (1 + 2*6 harvests excluding the last harvest) for the 21 years (Figure 1.b). Frequency of farm operations was in accordance with Hamelin *et al.* (2012). The average annual fertilizer input estimated from the life cycle years was comparable with Pugesgaard et al. (2015). Willow harvesting was assumed to occur every three years (i.e. a total of seven cuts), with the first harvest occurring after four years (Heller et al., 2003; Pugesgaard et al., 2015). The annual average yield was based on the studies reported by Hamelin et al. (2012) and Lærke *et al.* (2010) (Table 1). A single-stage harvester (cut and chip) with a fuel consumption of 14 lha^{-1} was assumed as the method of harvesting, which was representative to Danish practice (Djomo *et al.*, 2015). The fuel consumption was also consistent with the studies Goglio and Owende (2009) and Heller et al. (2003). The restoration process involved pressing back the stools into the soil and application of herbicides during summer (Gonzalez-Garcia *et al.*, 2012). Fuel consumption related to the pressing of stools was estimated to 38.7 l/ha (Njakou Djomo, 2016.pers. comm.).

With regard to alfalfa, it was assumed to be a rotational crop with a three-year rotational cycle (Jørgensen et al., 2011) and with three harvests per year. The yield (Table 1) was taken from NaturErhvervstyrelsen (2015) and Møller *et al.* (2005b). The quantity of seeds was calculated from Jørgensen et al. (2011). The annual application of fertilizers was based on SEGES (2010). Frequencies of farm operations were based on Jørgensen et al. (2011). Types of herbicides and total doses over the crop production cycle were based on SEGES (2010) (see SI, Table S.4). After the land preparation and growing the crop, the harvesting process was followed by mowing, swathing, baling and loading of the fresh biomass (Jørgensen et al., 2011). The baled biomass was assumed to be transported to a distance of 3 km to the farm (Table 1).

2.3.2. Calculation of emissions related to SOC change

SOC change was calculated from the net C input to the soil. Net C input was the difference between the organic matter available to the soil from the selected crops and the reference crop. Spring barley (with 100% straw incorporated into soil) was set as the reference crop (Table 2). Spring barley was considered as the reference crop as it is one of the marginal crop in Denmark potentially being displaced with the changes in the demand of land by other crops (Tonini et al., 2012). It should be noted that in this study, production of straw from spring barely is also one of the selected biomasses, which then will have no displacement as argued above. But, the production of straw, as one of the assessed feedstock accounted for 1 t DM recovered from the total yield from 1 ha of land. Rest of the residues was assumed to be ploughed back into the soil. The removed 1 t DM straw as feedstock to biorefinery was 46% of the total yield. This led to meet the sustainable rate of recovering straw from field. The sustainable recovery rate of straw is generally from 33% to 50% and was argued inducing marginal changes on the SOC (Scarlat et al., 2010; Spöttle et al., 2013).

The method to calculate the net C assimilation for spring barley (for straw) and alfalfa followed Taghizadeh-Toosi et al. (2014a) and was based on the non-harvestable above- and below-ground residues. The net non-harvestable residues for straw and alfalfa production were calculated from the parameter (i.e. ratio of the total DM available from stubbles and root to the net yield of the crops). The parameters for spring barley production was based on Taghizadeh-Toosi et al. (2014a). In the case of alfalfa the necessary parameters were calculated based on Djurhuus and Hansen (2003) and Pietsch *et al.* (2007) (see SI Table S.1).

In the case of willow, the non-harvestable above-ground biomass was partitioned into the DM yield from leaves and from woody material (branches, twigs), as shown in Eq. (i) (Hamelin, 2011). Likewise, the amount of below-ground residues was calculated from the fraction of total biomass production going to roots (f_R) using Eq. (ii) (Hamelin, 2011).

$$NHAG_{DMW} = \frac{f_{lw}}{f_{py}} \times PY + \frac{f_L}{(1-f_L-f_R)} \times \left[PY + \frac{f_{lw}}{f_{py}} \times PY \right]$$ ....Eq.(i) (Hamelin et al., 2012).

$$NHBG_{DMW} = \frac{f_R}{(1-f_L-f_R)} \times \left[PY + \frac{f_{lw}}{(1-f_{py})} \times PY \right]$$ ....Eq (ii) (Hamelin et al., 2012).

where $NHAG_{DMW}$ = non-harvestable above-ground DM for willow; f_{lw} = woody biomass loss during harvest = 7.5%; f_{py} = expected primary yield of the total potential primary yield (PY) = 92.5%; f_L = proportion of total biomass production going to leaves = 20%; f_R = proportion of total biomass production allocated to roots = 25%; $NHBG_{DMW}$ = non-harvestable below-ground residues for willow.

The primary yield (PY) of willow, i.e., the net biomass yield is shown in Table 1. All other assumptions on the partitioning of the non-harvestable biomass (f_{lw},f_L, f_{py}, f_R) , as shown in Eqs. (i and ii) are based on Hamelin et al. (2012).

Finally, SOC change for the selected biomasses production was calculated based on the respective net C input (Table 2). Emissions due to SOC change were calculated in a 100-year perspective, assuming an emission reduction potential of 9.7% of the net C input. Likewise, SOC change estimated for 20 years are also shown in Table 2, for which the emission reduction potential was set to 19.8% of the net C input to soil (Petersen *et al.*, 2013).

Table 1: Crop production data. All data are per ha

Table 2: Crop-specific assessment parameters used in the calculation of SOC change

2.3.3. Soil quality

There are number of factors that affect soil quality such as compaction, soil nutrients and SOC stock (Arshad and Martin, 2002). In this study, for the assessment of soil quality change in the SOC stock (Δ SOC, in t C hay^{-1}) was used as an indicator (Brandão et al., 2011; IPCC, 2000). The method used to calculate Δ SOC stock is presented in the form of Eq. (iii), and was in accordance with Brandão et al. (2011) and Milà i Canals et al. (2007b). The first component of the numerator in Eq. (iii) corresponds to the impact of the postponed relaxation of the land use system (i.e. during transformation), and the second component refers to the impact from changes in soil carbon in the current land use (i.e. during the occupation of the land) (Brandão et al., 2011). Relaxation was defined as the tendency of the soil quality of the current land use reverting to the prior level in terms of achieving the SOC stock of the reference situation (Brandão et al., 2011). For this purpose, natural forest can be regarded as a reference situation, assuming the current crop management was not in practice (Milà i Canals et al., 2007b). In this study Danish forestry was assumed as the reference situation, and the relaxation rate was adapted from Nielsen *et al.* (2010) and Grüneberg *et al.* (2014) (Table 3). Relaxation rate is the rate of SOC change that would take place if there is no transformation of the land use, or if the land was undisturbed. Relaxation time is another important parameter, since it is the period taken by the soil quality to revert to the equilibrium condition (Brandão et al., 2011). It was calculated from Eq. (iv). The calculation of Δ SOC stock was based on the amortization period of 20 years, where final year (t_f) = 20 years, initial (t_{ini}) = 19. The annualized change in SOC stock (Δ SOC stock, expressed as t C ha^{-1} y^{-1}) (Brandão et al., 2011) was thus calculated for the accounting period of 20 years. The temporal scope of 20 years was chosen to be consistent with IPCC (2000) for the assessment of soil quality. The Δ SOC stock was calculated considering the: (i) potential SOC stock (SOC_{pot}), i.e. if the forest land use was left undisturbed, (ii) initial SOC stock (SOC_{ini}), i.e. of the currently used arable land and (iii) final SOC stock (SOC_{fin}), i.e. the stock available at the end after the annual SOC change during the land occupation contributes to the SOC_{ini} (Table 7). Both SOC_{pot} and SOC_{ini} are shown in Table 3. The annual rate of SOC change due to the land occupation for producing the selected biomasses is shown in Table 2 (i.e. the values for 20 years).

$$\Delta SOCstock = \frac{(SOC_{pot}-SOC_{ini})*(t_{relax}-t_{ini})+1/2(t_{relax}-t_{ini})*(SOC_{ini}-SOC_{fin})}{(t_{fin}-t_{ini})}$$

.............Eq. (iii) (Brandão et al., 2011).

$$t_{relax} = \left[t_{fin} + \left(\frac{soilCchange}{relaxationrate} \right) \right] \qquad \text{.................Eq. (iv)}$$

Table 3: Basic parameters used for calculating the SOC stock change

2.3.4. Calculation of emission related to fertilizer application

A field N-balance method (Brentrup *et al.*, 2000; Hansen *et al.*, 2000) was used to calculate N-leaching. All the N-related inputs and outputs (e.g. plant uptakes) and losses (Table 4) were estimated before calculating N-leaching for the selected biomasses. Direct and indirect nitrous-oxide emissions (N_2O-N) were based on the emission factors reported in IPCC (2006). The emission factor for NH_3 emission was set to 2% of the N-fertilizer input (EEA, 2013; Nemecek and Kägi, 2007) and from the crops it was set to 0.5 kg N $ha^{-1}y^{-1}$ (Sommer *et al.*, 2004). Denitrification was calculated using the SimDen model (Vinther, 2005). These methods and models were used as they can represent variables of the specific agro-climatic condition that the current study has considered. The soil organic nitrogen (SON) change was calculated from the SOC change related to the land occupation of selected crops (Table 2) and applying the C/N ratio of 1:10. The method was in accordance with Mogensen et al. (2014).

Phosphorus (P) losses were set to 5% of the P-surplus (Nielsen and Wenzel, 2007). The P-surplus was calculated after accounting the P-uptakes by the plants (Møller *et al.*, 2000), as summarized in Parajuli et al. (2016).

Table 4: Biomass-specific N balances and emissions. All data are per ha

2.3.5. Calculation of freshwater ecotoxicity

The total PFWTox was calculated covering the two levels: (i) at farm level, estimating the emissions from pesticides during their application and (ii) at background level by covering the toxic emissions related to the production of the assumed material inputs entering into the agricultural system. At the farm level emission distribution fractions to air and freshwater of the respective active ingredients were calculated from the SI provided in Parajuli et al. (2016). The average emission distribution to air (first number in parentheses) and freshwater (second number in parentheses) were in the order of: herbicides (8%, 0.003%); fungicides (14.83%, 0.0003%); insecticides (5.63%, 0.00021%); growth regulator (36.92%, 0006%). For the calculation of total PFWTox, the chemical class of the pesticides was identified based on Footprint PPDB (2011) and ChemicalBook Inc. (2008), and when pesticide classes could not be identified from the two data sources they were classified as "unspecified class" (Weidema et al., 2013).

2.4. Sensitivity analysis

The sensitivity analyses covered following assessments and the results are presented in Table 7.

i. Variations in SOC change: It was calculated by varying the method to calculate the SOC change compared to the basic scenario (Table 2, values for 20 years). In the sensitivity analysis, the SOC change in 20 years was calculated using the method of IPCC Tier 1 (IPCC, 2006). The land use change factors assumed for the assessment are presented in SI Table S.2.

ii. Variations on the results due to changes in the assumptions. The assessment included:
   a. Effect of SOC change on $GWP_{100:}$ It included assessment of GWP without SOC change.
   b. Effect of different type of N-fertilizer: It included urea instead of Calcium Ammonium Nitrate (CAN) as a source of synthetic N-fertilizer. Changes were calculated for GWP_{100} and NRE use.
   c. Two-stage harvest of willow and effect on GWP_{100}: Specific fuel consumption in the two-stage harvest technology is presented in the foot notes of Table 7.

iii. Variation in soil quality: This included the assessment of soil quality by varying: (a) the rate of SOC change during the land occupation, as calculated from the above mentioned method (in the point ' i') compared to those presented in Table 2, and (b) the initial SOC stock (Table 5).

Table 5: Main parameters for the sensitivity analysis on the calculation of Δ SOC stock for the production of the selected crops

3. Results

3.1. Potential environmental impacts

Global Warming Potential: The obtained GWP_{100} for producing straw was 264 kg CO_2 eq tDM^{-1}. The impact of producing alfalfa and willow was only 32% and 38% respectively of the impact calculated for straw (Table 6). On a hectare basis the results are shown in Figure 2, which showed the lowest impact was for producing straw and the highest was for willow. In the case of producing straw, emission from SOC change contributed 17% of the impact. In contrast, for the production of willow and alfalfa the obtained SOC change was mitigating, respectively 66% and 44% of the impact. The contribution from N_2O emissions to the obtained GWP_{100} of straw, willow and alfalfa production was 32%, 37% and 16% respectively (Figure 3a). Variations on the N_2O emission was primarily due to different fertilization rate (see Table 4). The production of agro-chemicals contributed with 29%, 71% and 41% to the obtained impact for producing straw, willow and alfalfa respectively. The field operation processes (tillage, application of agro-chemicals and harvest) contributed with 17% for straw. For willow and alfalfa it was, respectively 45% and 75% of the obtained GWP_{100} (Figure 3a). Compared to other biomasses, alfalfa had higher contribution from the farm operation, which was partly due to higher frequency of harvesting and loading and also due to higher primary energy input to handle the biomass with higher moisture content (Table 1). The production of the willow cuttings contributed 4.4% to the total GHG emissions obtained for the biomass production. Contribution from the transportation was about 11% of the respective GHG emissions obtained for both willow and alfalfa, and was 2% for straw. With regard to the impact assessed per energy content of the biomass, it was lowest for willow, followed by alfalfa and straw (Table 6).

Eutrophication Potential: The eutrophication potential expressed per t DM of the biomass production was lowest for willow, followed by alfalfa and straw (Table 6). On a hectare basis EP was lowest for straw among the selected biomasses (Figure 2). The impact was primarily related to field emissions, e.g., nitrate leaching and ammonia and phosphate emissions (see related emissions in Table 4). These jointly contributed 40%, 46% and 68%, respectively of the total EP obtained for willow, alfalfa and straw (Figure 3b). It should be noted that emission factors to the EP are higher for NH_3, and N_2O emissions than nitrate emissions (Environdec, 2013), hence alfalfa with no synthetic fertilizer use had null NH_3 emissions (related to fertilizer) and lower N_2O emissions (Table 4) resulted with a lower EP compared to straw based on spring barley.

Non-Renewable Energy use: The obtained NRE use was highest for alfalfa, which was partly because of its higher harvesting frequency and higher primary energy use for baling the fresh biomass with higher moisture content (Table 1). On a hectare basis, NRE use was lowest for straw and highest for alfalfa (Figure 2). A major contributor to NRE use was the production of agrochemicals. Production of agro-chemicals contributed 20%, 45% and 47% of the total NRE use obtained for alfalfa, willow and straw productions respectively. For willow and straw the impact was mainly due to the production of N-fertilizer (Figure 3c). In contrast to the impact expressed per t DM, in terms of energy content it was the lowest for willow compared to the other biomasses. Production of willow cuttings contributed with 3% to the total NRE use calculated for willow, which was comparable to the range reported in Djomo *et al.* (2011).

Agricultural Land Occupation and Potential Freshwater Ecotoxicity:

The ALO was lowest for alfalfa, followed by straw and willow. With regard to PFWTox, particularly at farm level it was highest for straw, followed by alfalfa and willow (Table 6).

The total PFWTox also resulted to be higher for straw production from spring barley, and was lower in alfalfa and willow (Table 6). On the hectare basis, the impact was lowest for straw and highest for alfalfa.

Soil quality:

A detrimental effect to soil quality was found for straw compared to willow and alfalfa (Table 6), which was partly due to:

(i) differences in the SOC change during the land occupation: SOC change during the production of willow and alfalfa were -0.39 and -0.25 t C $ha^{-1}y^{-1}$ respectively (Table 2). In contrast emissions from the SOC change during the production of barley were 0.298 t C $ha^{-1}y^{-1}$ (Table 2).

(ii) higher difference between the relaxation rate and the SOC change: The relaxation rate was much higher (-0.31 t C $ha^{-1}y^{-1}$, Table 3) than the SOC change (0.298 t C $ha^{-1}y^{-1}$ in Table 2) in the case of producing spring barley. This thus requires a longer time to revert the soil quality to the prior situation (i.e. to the level of SOC_{pot}).

(iii) larger difference between the SOC_{pot} and SOC_{fin}: The impact was mainly caused by the postponed relaxation-time during the production of the selected crops depending on the differences between the SOC_{pot} with SOC_{ini} and SOC_{fin} (Table 3, Table 5 and Table 7). The rate of SOC change due to the land occupation (Table 2), as discussed above (in the point i) was the key factor on the scale of the differences between the SOC_{ini} and SOC_{fin}. Due to these differences the calculated relaxation time for spring barley was 20.96 years (Table 7), indicating that a longer period would be required to return to the level of natural relaxation (with forest as land use). A similar situation was for alfalfa, but the difference between the relaxation rate and the SOC change was not so high compared to spring barley. For willow there was an increase in the SOC stock, as the relaxation time was shorter (i.e. 18.73 years, see Table 7), hence soil quality would be able to revert quickly back to the reference situation (Table 7). Relaxatation time for the selected biomasses is shown in Table 7.

For an annual crop similar effect was reported in Brandão et al. (2011), and highlighted that a delay in relaxation would take place in such a situation (during the land use transformation) and hence land occupation itself has little effect compared to the delayed relaxation. The tendency of varying the soil quality as a result of different SOC change is further discussed in section 4, and the results are presented in Table 9.

Table 6: Environmental impact potentials per t DM biomass production

Table 7: Soil quality effects at the cropping stage

Figure 2: Environmental impact potentials per ha of the biomass production.

Figure 3: Environmental hotspots related to GWP_{100}, EP and NRE use.

4. Sensitivity analysis

Table 7 lists the variations in the results for the selected categories. Details on the specific assessments are as follows:

4.1 Variations in SOC change

With the use of IPCC method (IPCC, 2000), the annualized SOC change (in 20 years) for willow changed from -0.4 t C $ha^{-1}y^{-1}$ (basic scenario, Table 2) to -0.9 t C $ha^{-1}y^{-1}$ (Table 7). The result was however comparable to the range reported for SRC (Brandão et al., 2011; Dawson and Smith, 2007; Murphy et al., 2014). For alfalfa, the SOC change was -0.25 t C $ha^{-1}y^{-1}$ in the basic scenario (Table 2), which increased to -0.62 t C $ha^{-1}y^{-1}$ (Table 7) with the IPCC method. The range covering both methods was close to the reported values for perennial grasses and ley rotations, i.e. -0.5 to -0.62 t C $ha^{-1}y^{-1}$ (Dawson and Smith, 2007). In the case of straw it varied from 0.15 t C $ha^{-1}y^{-1}$ (Table 2) to 0.32 t C $ha^{-1}y^{-1}$ (Table 7).

4.2. Variations in the Global Warming Potential-100

a. Effect of SOC change on GWP_{100}*:* The carbon footprint of straw was 83% lower without SOC change, whilst it was 60% and 70% higher for willow and alfalfa (Table 6 and Table 8).

b. Effect of different type of N-fertilizer: Compared to CAN, if urea was assumed as the source of N-fertilizer then the carbon footprint of producing straw and willow would be lower by approximately 75%, but NRE use would be 94% higher (Table 6 and Table 8). The reason for the higher GHG emissions on the use of CAN was related to the emissions during nitric acid production, which is one of the important formulating compounds in the production of CAN (Agri-footprint, 2014). There are additional consequences of applying urea, which was not covered in the current assessment, e.g. higher NH_3 emissions. Such variation is primarily related due to how quickly plant can uptake the available N, which is in fact rely on the forms of available N from urea and CAN. This is however also relying on the agro-climatic conditions and the seasons of application of the fertilizers (Brentrup et al., 2000).

c. Two-stage harvest of willow and effect on GWP_{100}: A two-stage harvesting process was found to increase the obtained GWP_{100} and NRE use by 19% and 37%, respectively compared to the basic scenario (Table 6 and Table 8). This was due to the higher diesel consumption in the two-stage harvesting method (reported in the footnotes of Table 8).

Table 8: Sensitivity analysis on SOC change, GHG emissions and NRE use for the production of the selected biomasses compared to the basic scenario

4.3. Soil quality

Scenario-1 (S_1) analyzed the effect of different SOC change on the soil quality (Table 9). Hence, based on the SOC change estimated from the IPCC Tier 1 method, the change in SOC stock was 0.3, -1.44 and -0.76 t C $ha^{-1}y^{-1}$ for straw, willow and alfalfa, respectively. Since, the SOC change in S_1 was higher than the basic scenario, the differences between the SOC_{ini} stock and the SOC_{pot} stock was lower or more accumulation of SOC to the pool. This was the principal reason for the quick recovery of soil quality to the prior level, particularly for willow and alfalfa (Table 9). Furthermore, selection of the SOC_{ini} stock would also vary the results (Table 9). For instance, for all the biomasses, the difference between the SOC_{pot} and SOC_{fin} would be lower if higher values are selected for the SOC_{ini} (Table 5). In the case of alfalfa when the result of basic scenario was compared with the sensitivity scenarios (S1, S2 and S3), it can be concluded that SOC change induced during the land occupation was one of the determining factors to either revert the soil quality to reference situation by accumulating the SOC to the pool or deplete the quality by depleting the SOC input to the soil pool.

Table 9: Variations in calculated soil quality as a result of SOC change and initial SOC stock (values are given per ha; negative value indicates an increase in SOC stock)

5. Discussions

5.1. Comparing the selected environmental impact categories with other studies

5.1.1. Straw production

Mogensen et al. (2014) reported a carbon footprint for the production of straw from barley, excluding and including the SOC change to be 68 and 91 kg CO_2 eq t DM^{-1}, respectively. The difference in the carbon footprint compared to our study was partly due to the use of different allocation factors (5% of the grain in their study), fertilization rates and emission factors of diesel use and fertilizer production. In addition, there were also differences in the estimates on the SOC change. In contrast, Korsaeth *et al.* (2012) reported a carbon footprint of straw from spring barley as 356 kg CO_2 eq t DM^{-1} (with SOC changes), which nominally differed from this study and was mainly due to different allocation factors. Despite the tools used to calculate SOC change were different but was based on similar approaches. Although there were variations in the results compared to other studies, based on the contribution from biomass production value chains the results were comparable with the stated other studies. For instance, the contribution of N_2O emissions to the GWP_{100}, as reported in this study (section 3.1) was found to be similar to the range reported in Roer *et al.* (2012) and Kramer *et al.* (1999).

Niero *et al.* (2015), Roer et al. (2012) and Korsaeth et al. (2012) reported a higher equivalent score for freshwater ecotoxicity for spring barley compared to this study. The reason behind the differences was partly due to the different types and amount of pesticides applied, and apparently a dissimilar emission distribution fractions of applied pesticides might be principal reason for the differences. Furthermore, in Niero et al. (2015) emissions from the inorganic elements deriving from animal slurry was also included, which was one of the main reason for the difference.

5.1.2. Willow production

The carbon footprint of SRC, including willow, ranged from 0.6-12 kg CO_2 eq GJ^{-1} (Djomo et al., 2011; Dubuisson and Sintzoff, 1998; Krzyzaniak *et al.*, 2013; Matthews, 2001; Murphy et al., 2014; Pacaldo *et al.*, 2012). Heller et al. (2003) reported a value of 0.68 kg CO_2 eq GJ^{-1}, its size explained by the higher carbon sequestration, which was based on below-ground residues. There were also some variations in the methods used to estimate the residues and carbon assimilation, e.g. with regard to the method for calculating the below-ground residues. For instance, the shoot-to-root ratio was used in Pacaldo et al. (2012) and Heller et al. (2003). Sartori *et al.* (2007) reported both decline and increase in the SOC change for the different methods used for calculating the available residues in soil. Brandão et al. (2011) reported farm-gate GHG emissions was -102 kg CO_2eq GJ^{-1} (with -497 kg CO_2 eq $ha^{-1}y^{-1}$ avoided due to SOC change), but excluding SOC change gave comparable results to the current study (Figure 2).

Diesel used in farm operations for willow contributed 0.5 GJ $ha^{-1}y^{-1}$ (Table 1) and was comparable to those found by Matthews (2001) and Pugesgaard et al. (2015). Including the background processes, the total NRE use calculated per ha (Figure 2) was also close to 21.3 GJ $ha^{-1}y^{-1}$, as reported by Matthews (2001). In contrast, Brandão et al. (2011) reported 6.4 GJ $ha^{-1}y^{-1}$ as the total energy input. Minor differences compared to our study were related to the processes covered by the background system, assumed life cycle span and the frequency of fertilization. Regarding the freshwater ecotoxicity calculated for the foreground system it was comparable to that of *Salix* (Nordborg *et al.*, 2014).

5.1.3. Alfalfa production

Alfalfa production, as undersown in rotation (corn-soybean-alfalfa, conventional) was reported with GHG emission and NRE use as 71 kg CO_2 eq ha^{-1} $y^{-1}$and 1.5 GJ ha^{-1} respectively (Adler *et al.*, 2007). The differences in the results were also partly due to different emission factors assumed for diesel use and the different system boundary used for the assessment. In contrast, Gallego et al. (2011) reported a higher carbon footprint and a total NRE use of 3.8 GJ t DM^{-1}. The difference was due to consideration of a drying process to achieve a higher DM content (i.e. 89%) in their study. If the drying process was excluded from their results, the value for NRE use was comparable. Likewise, Sooriya Arachchilage (2011) and Vellinga *et al.* (2013) reported that GHG emissions for alfalfa (including the transportation to biorefinery plant) was about 100 kg CO_2 eq t DM^{-1} including transport to a biorefinery plant, which was close to our result. The reported NRE use by Vadas *et al.* (2008) was 4 GJ ha^{-1}, and this was based on the mass allocation from the total normal yields of crops in a four-year rotation. The results of the current study on ha basis are shown in Figure 2.

With regard to EP, values for alfalfa ranged from 0.4 to 1.14 kg PO_4^{3-}eq t DM^{-1} (Gallego et al., 2011; Sooriya Arachchilage, 2011). The major contributing processes and emissions were from applied N fertilizer, and the main substances responsible for the impact were NH_3 emissions, nitrate and phosphate leaching, which is consistent with the results of the current study, as reported in Table 4.

5.2. Soil quality and the affecting factors

In this study, an accumulation of SOC was found during the production of willow (i.e. -1.06 t C ha^{-1} y^{-1}, Table 9), which was the result of a higher SOC change (Table 2) relative to the relaxation rate (Table 5). The annualized SOC stock change (in t C ha^{-1} y^{-1}) for SRC is reported to range from -0.3 to -2.8 t C ha^{-1} y^{-1}, depending on the annualized period used for the calculation (e.g. 25 to 115 years) (Dawson and Smith, 2007). The results obtained in our study also fell within that range, as did the results of Falloon *et al.* (2004) and Murty *et al.* (2002). Willow showed high potential for a quick recovery due to higher SOC change during the land occupation than the relaxation rate (Table 9). This was opposite in the case of alfalfa, but it was varying with the different rate of SOC change, as discussed in section 4.3 (Table 9). For instance, for alfalfa potential improvement to the soil quality was found in S_1 and S_3 compared to the basic scenario and S_2 (Table 9). Likewise, the annualized Δ SOC stock for alfalfa (Table 6 and Table 9) was found comparable to leys in rotation and permanent grassland (-0.35 to -1.6 t Cha^{-1} y^{-1}), as reported in Guo and Gifford (2002), Murty et al. (2002) and Smith *et al.* (1997). Termansen *et al.* (2015) reported that the effect on SOC stock during the shift from a cereal crop rotation to grass was about -0.49 t C ha^{-1}y^{-1}in Danish soil, and further argued that it will take place over a longer period until a new equilibrium in the soil is reached (estimated to be 20-40 years). This was comparable to the situation for alfalfa, as reported under S1 in the sensitivity analysis (Table 9). Meanwhile, there was a depletion of SOC stock in the case of spring barely production (Table 6 and Table 9).

In general, conversion of a natural ecosystem, such as forest and grassland to managed agriculture has about 10-59% decline in SOC stock. On the other hand by replacing crops with pasture and woody plantation tends to increase SOC stock (Qin *et al.*, 2016). In this study, based on the obtained final SOC stock the impact of land use conversion (i.e. from forest to arable land) showed 54% decline in SOC stock (Table 7 and Table 9). Moreover, in relative to the initial SOC stock, the final SOC stock for willow and alfalfa showed accumulation of SOC by 0.44% and 0.28% respectively, whilst the depletion in the case of spring barley was 0.33%. Tonini and Astrup (2012) reported that during a land use change from spring barley to willow the SOC stock change was -15 t Cha^{-1}; and during the conversion from cropland to grassland it was -8 t C/ha. This was comparable to the non-annualized values of willow, i.e. -21 t C ha^{-1} and -29 t C ha^{-1}, as calculated from the basic scenario and S1 (Table 9). For alfalfa, based on S_1 it would be -15 t Cha^{-1} (calculated from Table 9).

5.3. Utilization of biomasses

Based on the energy content of the selected biomasses, the current study showed that for most of the impact categories, willow performed better compared to the rest of the biomasses (Table 6). In addition, the total energy output-to-input ratio for producing 1 t DM of biomass was 7, 13 and 7 for straw, willow and alfalfa, respectively. The value for willow was close to the ratio of SRC reported in Manzone *et al.* (2009) and also corresponds to the lower range for SRC reported in Djomo et al. (2011). The energy output to input ratio are relevant when the biomasses have to be considered for thermo-chemical conversion of biomasses (McKendry, 2002a). Moreover, other physio-chemical compositions of the biomass are also relevant to prioritize them for specific biorefinery platforms (Parajuli et al., 2015b). For instance, carbohydrate content of alfalfa, willow and straw are 60%, 56% and 76% (Møller *et al.*, 2005a; Parajuli et al., 2015b). On the basis of carbohydrate content of straw, the equivalent mass of alfalfa and willow would be thus 1.18 and 1.1 t DM. Hence the environmental impacts of their biochemical conversions (e.g. in sugar based platform of biorefinery) (Parajuli et al., 2015b) would be therefore differing based on the conversion efficiency of the carbohydrates in the related biorefinery processes (Huang and Zhang, 2011). Likewise, in general, net bio-energy conversion efficiencies for biomass combustion in power plants range from 20% to 40%, integration of gasification and combustion (40-50%), pyrolysis to produce bio-oil (up to 80%) (McKendry, 2002b) and for conversion to bioethanol up to 70% (Larsen and Henriksen, 2014). In addition to these, if biomass utilization efficiency (BUE) is used as a proxy indicator to measure the efficiency of utilizing waste produced during their conversions then the conversion of biomass to bio-methane showed BUE as 20.3, bioethanol (47.2 from glucose, whilst 34.6 to 38.1 from cellulose), pyrolytic gasification (12.1 from cellulose), biodiesel (72.7 to 98) (Iffland et al., 2015). These showed that optimum utilization of resources would thus be beneficial for their sustainable conversions.

5.4. Consequences of biomass utilization

For the sustainability of biorefinery and bioenergy value chains the most important aspect is to maintain a year round supply of biomass. Hence, this stresses to assess potential consequences of utilizing biomasses, e.g. in relative to the current applications. For instance, the current application sides of Danish recovered straw are 49% as fuel, 32% for fodder, and 19% as bedding materials in livestock houses (Gylling *et al.*, 2013). Likewise, alfalfa is used as a fodder and bioenergy crops (Sørensen *et al.*, 2013). Willow is also increasingly used as one of the options to energy crops (Nord-Larsen *et al.*, 2015). Here, potential consequences would be therefore on SOC change and soil fertility, if exploitation of residues exceeds the sustainable recovery rate (Scarlat *et al.*, 2010). Likewise, balancing the supply and demand of biomass both as bioenergy crops and biorefinery feedstocks would also be pertinent to examine in the transitions of biomass applications (Parajuli et al., 2015a). On the other hand, it was argued that biorefineries will be able to produce animal fodder, which can replace some of the cereal that is used for animal fodder today. Estimates showed that if 10-15 % of the dry matter in straw and grasses is converted to animal feed, a comparable feed production will be able to achieve to what it is lost from the smaller area with cereal and rape (Gylling et al., 2013). These features revealed that over-exploitation of biomasses for energy purpose or for the production of materials could be an issue, in the absence of proper management of land use, and have to be taken seriously if these biomasses are going to be a fundamental platform of a Danish bioeconomy (Parajuli et al., 2015a). On the other hand, it was also revealed that the opportunities of co-producing different products from biorefineries can partially check the potential competitions among their alternative applications.

Apart from above discussed issues, in the current study effects of indirect land use change was not included in the assessment of GHG emissions. It was to avoid methodological chaos, which can be caused by summing average and marginal effects (Creutzig *et al.*, 2012). These however can be diligently examined when evaluating conversion of biomass into different biobased products by adopting different approaches of the LCA.

6. Conclusions

The general conclusion of the study was that the advantages of perennial crops over annual crops were their higher dry matter and energy yield, and were with relatively lower potential environmental impacts. Net biomass yield was the driving factor for lowering the environmental impacts for willow and alfalfa compared to straw. This was revealed from the differences on the results presented on hectare basis and per t DM basis for the selected crops. The impact was also determined by the material inputs, e.g. synthetic fertilizers, mainly N-fertilizer, types and amount of pesticides and the frequency of farm operations assumed during the production of the selected crops.

The study also showed the importance of understanding the implications of different agricultural management practices to the overall environmental impact potentials, for example with regard to SOC changes, maintenance of soil health and emissions from field operations. Willow and alfalfa contributed positively to soil quality, and the result was depending on the rate of SOC change that is induced during the land occupation. Willow and alfalfa had a higher nutrient use efficiency and lower nutrient leaching, thus had relatively lower EP. In addition, this study also showed that N_2O emission was one of the major contributors to GWP_{100} obtained for the respective biomasses. For almost all impact categories the production of agro-chemicals had the largest impact. This stresses the need of minimizing the use of synthetic fertilizer, e.g. by recycling/reusing organic matter in waste streams of biomass conversion technologies such as biorefineries. In the context of diversifying the biomass supply, particularly in the thermo-chemical conversion routes it is relevant to know if the biomass production system is a net energy producer or a consumer. On such assessment on willow showed it performing better among the selected biomasses. With regard to NRE use, straw had the lowest impact compared to the rest of the biomasses. The agricultural land occupation was lowest for alfalfa followed by straw and was highest for willow. These showed mixed results for the biomasses with regard to different environmental impact categories.

Finally, a comparison of biomass feedstocks as assessed at the farming system level may not give a complete picture of the environmental sustainability, as it also depends on how feedstocks are going to be utilized to satisfy societal demands. Feedstocks are also dependent on their chemical constituents and hence their conversion efficiency in bioenergy and biorefinery production chains. Hence, a future research perspective could be to assess the environmental and economic impact of biomass conversions in relevant biorefinery platforms and compare them with the conventional products. This requires integration of an agricultural system LCA, e.g. assessed at the farm gate level as in this study, with the LCA of the industrial processing of biomass to produce biobased products, e.g., via a biorefinery.

Acknowledgement

The article is written as part of a PhD study at the Department of Agroecology, Aarhus University (AU), Denmark. The study is co-funded by the Bio-Value Platform (http://biovalue.dk/), funded under the SPIR initiative by The Danish Council for Strategic Research and The Danish Council for Technology and Innovation, case no: 0603-00522B and is moreover relevant to the Nitroportugal EU project. The first author would like to thank the Graduate School of Science and Technology (GSST) of AU for the PhD scholarship. Thanks to Margit Schacht (from Agro Business Park) for providing necessary support in editing this article. We appreciate the comments provided by the independent reviewers that helped to improve the paper.

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Figure captions

Figure 1: System boundaries for the selected biomasses and related elementary flows. (Figure 1a represents the general system boundary and Figure 1b represents the production cycle of willow.).

Figure 2: Environmental impact potentials per ha of the biomass production.

Figure 3: Environmental hotspots related to GWP_{100}, EP and NRE use.

Table 1: Crop production data. All data are per ha

Materials	Unit	Amount			Remarks
		Spring barley-straw	Willow	Alfalfa	
Inputs					
Land (ha)	ha	1	1	1	
Seed (kg)	ha^{-1}y^{-1}	32	-	11	(Jørgensen et al., 2011).
Cuttings	numbers ha^{-1}	-	12000	-	See section 2.3.1
Synthetic fertilizer[a]	kg ha^{-1}y-^{1}				(NaturErhvervstyrelsen, 2015)
N		23	74[b]	-	
P		6	32	33	
K		8	172	214	
Lime	kg ha^{-1}y^{-1}	31.7	8	56	after Hamelin et al. (2012)
Pesticides	kg ha^{-1}y^{-1}	0.11	1.04	0.33	SI (Table S.5)
Lubrication oil	l ha^{-1}y^{-1}	2	4	14	Dalgaard *et al.* (2001)
Direct primary energy input	MJ ha^{-1}y^{-1}	492	458	4189	diesel (a + b); cuttings included in the case of willow (SI Table S.3).
a. Field preparation[b]	MJ ha^{-1}y^{-1}	325	214	688	Diesel input (Dalgaard et al., 2001)
b. Harvesting + loading - handling[c]	MJ ha^{-1}y^{-1}	167	234	3501	
c. Transport					
- seeds[d]	t km ha^{-1}	6.1	-	2	
Cuttings	t km ha^{-1}	-	48	-	SI, Table S.3
- agrochemicals[e]	t km ha^{-1}	14.25	73	78	
- biomass	t km ha^{-1}	4.18	64	105	

(field to farm)[f]					
Output at farm gate (net yield)					
Dry matter yield	t DM ha^{-1} y^{-1}	2.24	10.63	12.2	See section 2.3.1
Lower heating value[g]	GJ ha^{-1} y^{-1}	34	199	170	

Assumptions:

[a] N-fertilizer input: N-norms —N-fixation + N-seeds + N-deposition. (see Table 4)

[b] Included tillage and application of agrochemicals. Heating value of diesel = 35.95 MJl^{-1}, Density = 0.84 kg/l (Weidema et al., 2013).

[c] Calculation for the loading and handling:

[†] Baling = DM/ha * bale/160 kgfw/% DM *1000 kg/t * 0.23 (Hamelin et al., 2012). Diesel input = 0.743 kg bale^{-1}.

[ρ] Bale loading (straw and alfalfa) = (Number of bales/ha /0.23) * 0.0811 kg/bale (Hamelin et al., 2012).

[↓] Loading for barley grain = 0.119 litre m^{-3} fodder (Møller et al., 2000). Fodder (m^3) = DM/ha * kgfw/DM% * 0.004 m^3 fodder loading/kgfw *1000 kg/t (Hamelin et al., 2012).

[d] Mass of seed * distance (= 200 km) (Parajuli *et al.*, 2014).

[e] Materials (fertilizer + lime + pesticides) * distance (200 km)

[f] Tonnes of fresh biomass (at farm) * 3 km (single trip). Distance assumed, as in Mogensen et al. (2014). DM content: straw (85%) and alfalfa (35%) (Møller et al., 2005b), willow (50%) (Heller et al., 2003). The emission stage for the truck used was EUR5 (Weidema et al., 2013), single trip.

[g] Lower heating value (MJ kgDM^{-1}): *straw bales = 15 (Nielsen, 2004); alfalfa bales = 14 (Jørgensen *et al.*, 2008); willow chips = 18.7 (Pugesgaard et al., 2015).

Table 2: Crop-specific assessment parameters used in the calculation of SOC change

Parameters/Crop types	Unit	Spring barley	Willow	Alfalfa
Net biomass yield[a]	t DM ha^{-1}y^{-1}	4.08	10.63	12.2
Straw yield	t DM ha^{-1}y^{-1}	(2.24)[±]	-	-
Plant growth, total	t DM ha^{-1}y^{-1}	10.44[b]	13.27[c]	22.7[b]
Below-ground residues[b]	t DM ha^{-1}y^{-1}	1.77[b]	5.22[c]	5.92[b]
Above-ground residues	t DM ha^{-1}y^{-1}	3.55[d]	5.46[c]	3.17[d]
Total plant residues[e]	t DM ha^{-1}y^{-1}	5.32	10.69	9.09
Plant residues N[f]	t N ha^{-1}y^{-1}	$4.5*10^{-2}$	$5.3*10^{-2}$	$8.9*10^{-2}$
C input from residues from the reference crop[g]	t C ha^{-1}y^{-1}	0.29	0.29	0.29
C input from DM from the selected crops[g]	t C ha^{-1}y^{-1}	1.4	4.92	4.2
SOC change				
- in 100 years[h]	t C ha^{-1}y^{-1}	0.15	-0.19	-0.12
- in 20 years[i]	t C ha^{-1}y^{-1}	0.3	-0.4	-0.25
Emissions from SOC change (100-years)[j]	t CO_2 ha^{-1}y^{-1}	0.54	-0.71	-0.45

Assumptions:

[±] Value in the parenthesis for spring barley represent the straw yield.

[a] See section 2.3.1 for the data on biomass yield.

[b] Calculated based on Harvest index (alpha) and root mass (beta) relative to above-ground residues for: barley (Taghizadeh-Toosi et al., 2014a); for alfalfa elaborated in SI, Table S.1. In the case of barley, 1 t DM straw (i.e. 46% of the straw yield) was removed from the field, as the feedstock.

[c] Non-harvestable residues of willow were calculated based on Eq.(i) and Eq. (ii).

[d] Non-harvestable above-ground residues = Total plant residues – total root residues.

[e] Total non-harvestable plant residues = above ground + below ground residues.

[f] Calculated from the "Total plant residue [d]". Norms of N content (% DM) in stubble/straw, root. CP = Barley (10.6, 3.3) (average of years 2000-2013, based on reports (Møller et al.,

2005a; Møller *et al.*, 2012; Møller and Sloth, 2013; Møller and Sloth, 2014; Vils and Sloth, 2003); willow (0.45) (Pugesgaard et al., 2015); and alfalfa (16.2, 14.7) (Djurhuus and Hansen, 2003; Thøgersen and Kjeldsen, 2014).

[g] Calculated from the total C assimilation, i.e. 46% of the DM input (Taghizadeh-Toosi et al., 2014a).

[h] SOC change in 100 years = 9.7% of net C input (Petersen et al., 2013). Negative values indicate soil C sequestration

[i] SOC change in 20 years = 19.% of net C input (Petersen et al., 2013). Negative values indicate soil C sequestration.

[j] Emission from SOC change (in t C ha^{-1}y^{-1}) multiplied by the ratio of the mol. weight of CO_2 to C (44/12).

Table 3: Basic parameters used for calculating the SOC stock change

Parameters	Basic Scenario
SOC change during the land occupation (t C ha^{-1} y^{-1})	See Table 2
Natural relaxation rate (t C ha^{-1} y^{-1})[a]	-0.31
SOC_{ini} stock (t C ha^{-1})[b]	90
SOC_{pot} stock (t C ha^{-1})[c]	168

Assumptions:

[a] Danish forest land was used as the reference situation and the relaxation rate was assumed as -0.31 t C/ha/y (Grüneberg et al., 2014; Nielsen et al., 2010). Negative value indicates soil C sequestration during the reference situation.

[b] SOC_{ini} stock of agricultural land (Taghizadeh-Toosi *et al.*, 2014b).

[c] SOC_{pot} stock based on forest land use (Krogh *et al.*, 2003).

Table 4: Biomass-specific N balances and emissions. All data are per ha

	Unit	Amount			Comments/Remarks
		Barley-Straw[†]	Willow	Alfalfa	
Total N-input[a]	kg N ha^{-1}y^{-1}	26	89	358	
N-output[b]	kg N ha^{-1}y^{-1}	16	48	291	Table 1
Field balance	kg N ha^{-1}y^{-1}	10	41	67	N_{input}-N_{output}
N losses	kg N ha^{-1}y^{-1}				
NH_3-N		0.83	3.49	0.5	(EEA, 2013; Nemecek and Kägi, 2007; Sommer et al., 2004)
NOx-N		0.11	0.48	0.07	NO_x-N: NH_3-N = 12:88 (Schmidt and Dalgaard, 2012)
Denitrification		0.17	9	13	(Vinther, 2005).
Soil change, N	kg N ha^{-1}y^{-1}	-3.61	19	13	See section 2.3.4
Potential leaching	kg N ha^{-1}y^{-1}	11	9	41	Field balance - losses
Total N_2O-N losses (direct +indirect)	kg N ha^{-1}y^{-1}	0.41	0.85	0.34	(IPCC, 2006)
P losses	kg P ha^{-1}y^{-1}	0.15	1.6	1.65	Section 2.3.4

Assumptions:

[†] N balance for straw was allocated from the spring barley production.

[a] Total N-input = $F_{SN} + N_{fixation}^{\rho} + N_{deposition}^{\dagger} + N_{seed}^{\pm}$.

[ρ] $N_{fixation}$ for alfalfa = 353 kg N ha^{-1}y^{-1}(Høgh-Jensen and Kristensen, 1995) and (Rasmussen *et al.*, 2012).

[†] N deposition = 15 kg N ha^{-1} (Ellermann *et al.*, 2005)

[±] N_{seed} calculated after the Farm-N model (Jørgensen *et al.*, 2005).

[b] Calculated based on Crude N and the DM yield. kg N per t DM yield for: spring barley = 0.0173 and straw= 0.006 (Møller et al., 2012; Møller and Sloth, 2013, 2014; Vils and Sloth, 2003)), alfalfa =0.024 (Møller et al., 2005a); Thøgersen and Kjeldsen (2015) and willow =

0.0045 (Pugesgaard et al., 2015).

Table 5: Main parameters for the sensitivity analysis on the calculation of Δ SOC stock for the production of the selected crops

Parameters and scenarios	Scenario 1 (S_1)	Scenario 2 (S_2)	Scenario 3 (S_3)
SOC change for the selected crops (t C ha^{-1} y^{-1})	IPCC Tier 1[a]	Table 2[b]	IPCC Tier 1[a]
Relaxation rate (t C ha^{-1} y^{-1})[c]	-0.31	-0.31	-.31
SOC_{ini} stock (t C ha^{-1})	153[d]	153[d]	140[e]
SOC_{pot} stock (t C ha^{-1} a)[e]	168	168	168

Assumptions:

[a], SOC change (in 20 years) based on IPCC method.

[b] Table 2 and using the (Petersen et al., 2013) method for 20 years.

[c] Relaxation rate = -0.31 t C ha^{-1} y^{-1}(Grüneberg et al., 2014; Nielsen et al., 2010). Negative values indicate soil C sequestration.

[d] Based on Adhikari *et al.* (2014).

[e] Based on Krogh et al. (2003).

Table 6: Environmental impact potentials per t DM biomass production

Environmental impacts	Unit	Spring barley-straw	Willow	Alfalfa
GWP_{100}				
- with SOC change[a]	kg CO_2 eq t DM^{-1}	264	100	84
	kg CO_2 eq GJ^{-1}	18	5	6
EP	kg PO_4 eq t DM^{-1}	1.35	0.8	1.26
	kg PO_4 eq GJ^{-1}	0.09	0.04	0.09
NRE use	MJ eq t DM^{-1}	1225	1416	1991
	MJ eq GJ^{-1}	82	76	143
ALO	m^2 t DM^{-1}	869	949	852
	m^2 GJ^{-1}	58	51	61
PFWTox				
- at field level only	CTU_e t DM^{-1}	33	0.35	4.44
	CTU_e GJ^{-1}	2.23	0.02	0.32
- total	CTU_e t DM^{-1}	113	61	71
	CTU_e GJ^{-1}	8	3	5
Soil quality (Δ SOC stock)[b]	t C t DM^{-1}	1.22	-0.1	0.06
	t C GJ^{-1}	0.08	-0.01	0.004

[a] SOC during the occupation of land.

[b] ΔSOC stock indicates the change in the SOC stock due to transformation and the occupation of land (see section 2.3.3). Negative value indicates an accumulation of SOC to the pool.

Table 7: Soil quality effects at the cropping stage

Biomass source	SOC_{ini} [a]	SOC_{fin} [a,b]	t_{relax} [a]
Spring barley	90	89.7	20.96
Willow	90	90.39	18.73
Alfalfa	90	90.25	19.2

[a] See section 2.3.3.

[b] $SOC_{fin} = SOC_{ini}$ + SOC change during the land occupation.

Table 8: Sensitivity analysis on SOC change, GHG emissions and NRE use for the production of the selected biomasses compared to the basic scenario

Impact potentials for the alternative scenarios	Spring barley straw	Willow	Alfalfa
A. Emissions due to soil C change in 20 years[a] (t C $ha^{-1}y^{-1}$)			
- Basic scenario [a]	0.3	-0.4	-0.25
- Based on IPPC Tier 1 method (IPCC, 2006)[b]	0.32	-0.9	-0.62
B. Net GWP_{100} (kg CO_2 eq t DM^{-1})			
i. with SOC change[c]	264	100	84
ii. without SOC change[d]	222	167	120
iii. Changed N-fertilizer use (Urea)[e]			
- Net GWP_{100} (kg CO_2 eq t DM^{-1})	212	63	-
- NRE use (MJ eq t DM^{-1})	1283	1486	-
iv. Use of two-stage harvesting method for willow[f]			
- Net GWP_{100} (kg CO_2 eq t DM^{-1})	-	119	-
- NRE use (MJ eq t DM^{-1})	-	194	-

Assumptions:

[a] See Table 2

[b] See SI, Table S.2 for the factors of the land use changes.

[c] See Table 6.

[d] Calculated from Table 6 by deducting the SOC change estimated for 100 years (see Table 2).

[e] Emission factor for Urea: GWP_{100} = 1.24 kg CO_2 eq kg N^{-1} and NRE use = 53.51 MJ eq kg N^{-1} (Agri-footprint, 2014).

[f] The basic scenario included singe stage harvester (cut and chip) (see section 2.3.1). For two-stage harvester, diesel consumption = 22 kg ha^{-1} (for cutting) and 21 kg ha^{-1} (for chipping) (Berhongaray *et al.*, 2013).

Table 9: Variations in calculated soil quality as a result of SOC change and initial SOC stock (values are given per ha; negative value indicates an increase in SOC stock)

Scenarios	Spring barley straw		Willow		Alfalfa	
	Δ SOC stock (t C $ha^{-1}y^{-1}$)	relaxation time (years)	Δ SOC stock (t C $ha^{-1}y^{-1}$)	relaxation time (years)	Δ SOC stock (t C $ha^{-1}y^{-1}$)	relaxation time (years)
Basic scenario[a]	1.47	20.96	-1.06	18.73	0.77	19.2
Sensitivity scenarios [b]						
S_1	0.30	21.03	-1.44	17.08	-0.76	18
S_2	0.29	20.96	-0.21	18.73	0.15	19.2
S_2	0.55	21.03	-2.69	17.08	-1.42	18

[a] Methods for the calculation are described in section 2.3.3.

[b] Scenarios for the sensitivity analysis and the parameters are shown in Table 5.

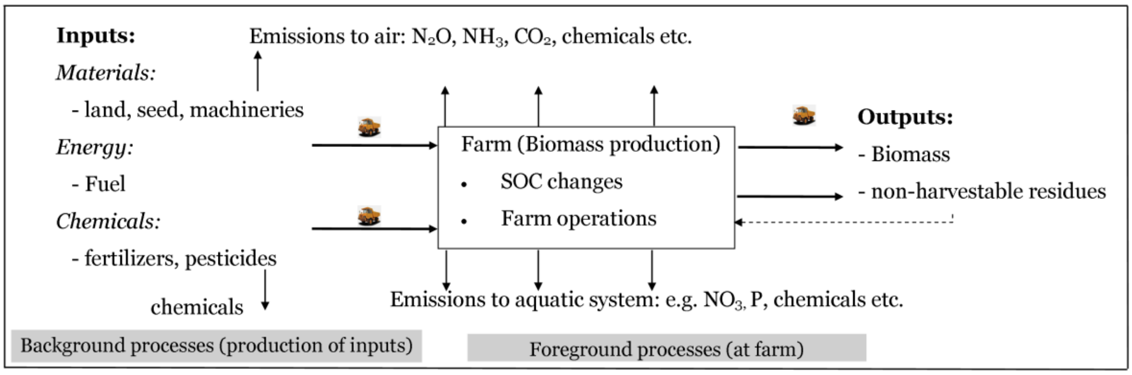

Figure 1.a. Schematic system boundary for the biomasses production

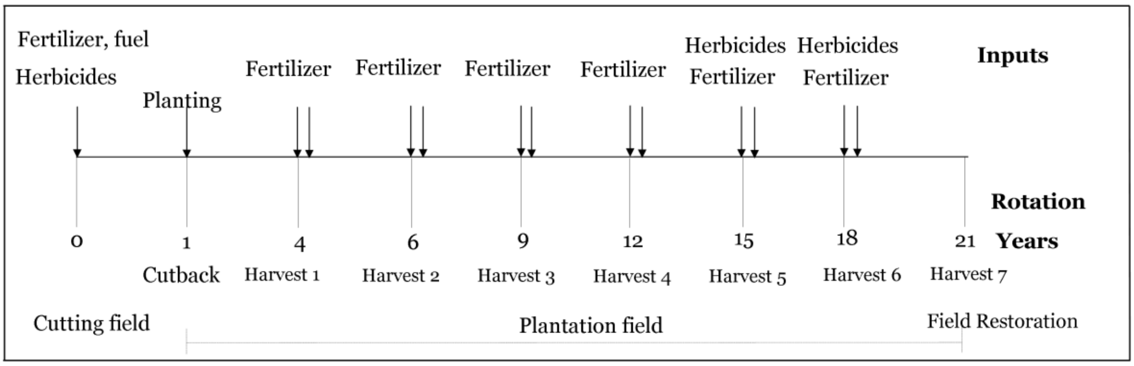

Figure 1.b. Schematic system boundary for the detail material inputs/activities for willow production at Foreground level

Fig. 1

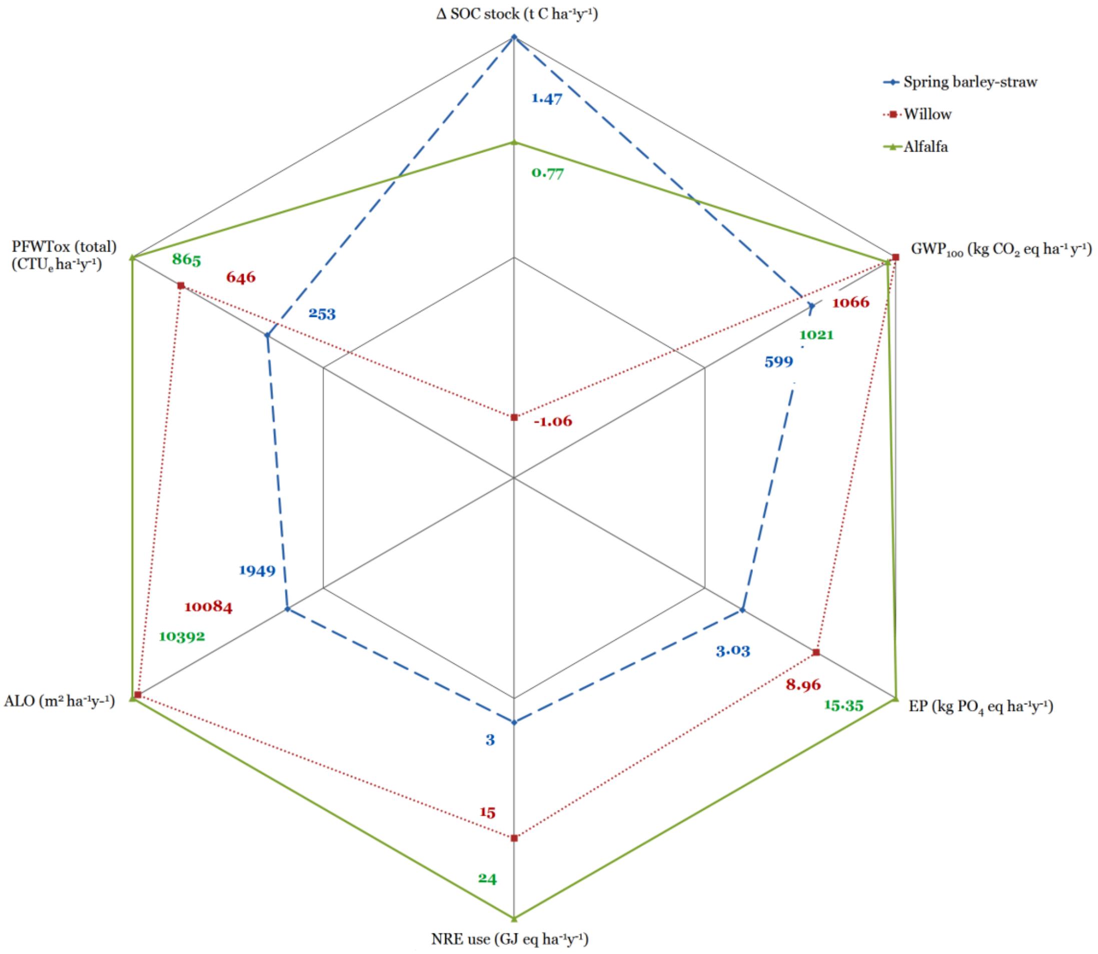
Δ SOC stock (t C $ha^{-1}y^{-1}$)
Spring barley-straw
Willow
Alfalfa
1.47
0.77
GWP$_{100}$ (kg CO_2 eq ha^{-1} y^{-1})
1066
1021
599
-1.06
PFWTox (total) (CTU_e $ha^{-1}y^{-1}$)
865
646
253
1949
10084
10392
ALO (m^2 $ha^{-1}y^{-1}$)
3.03
8.96
15.35
EP (kg PO_4 eq $ha^{-1}y^{-1}$)
3
15
24
NRE use (GJ eq $ha^{-1}y^{-1}$)

Fig. 2.

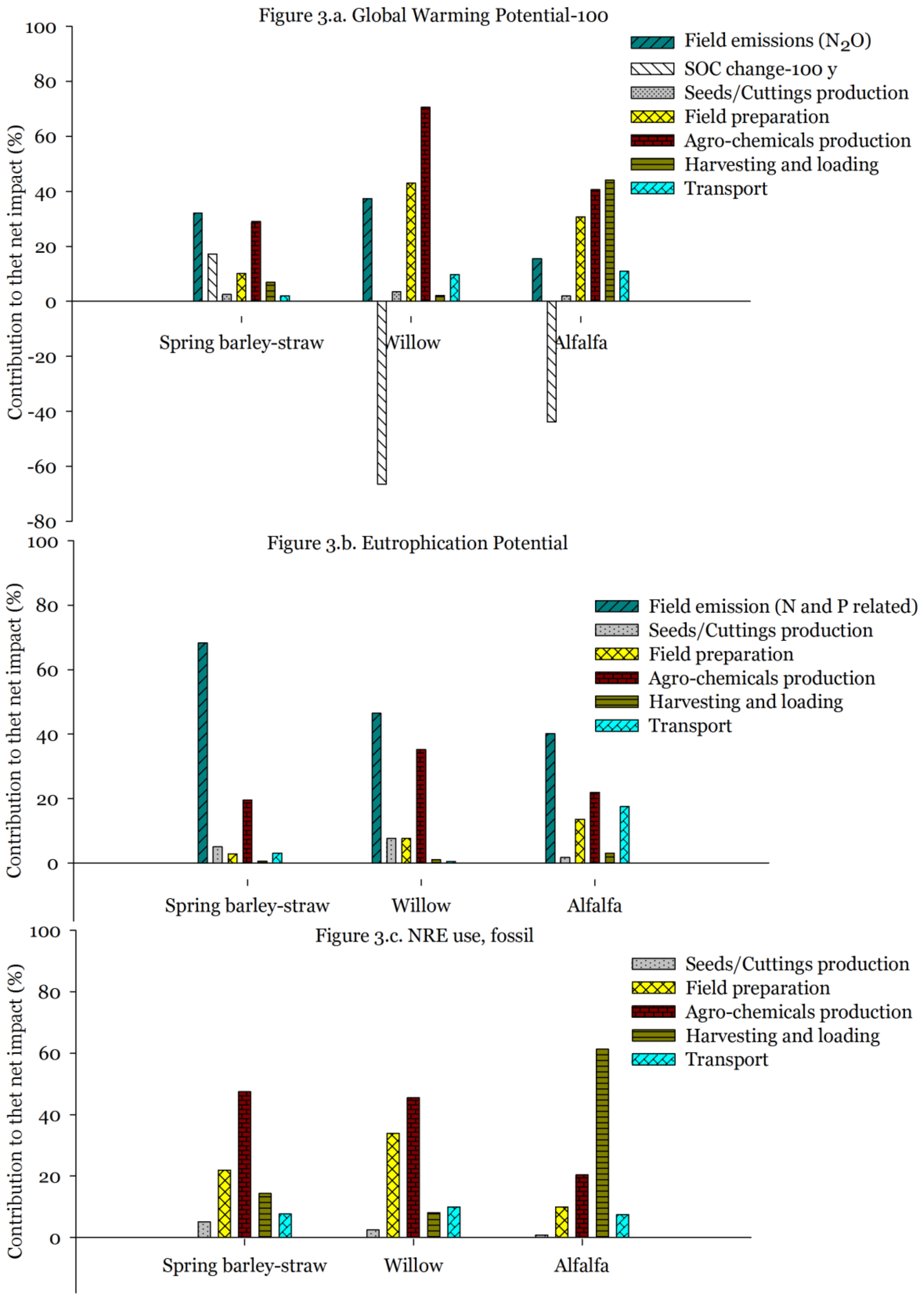
Figure 3.a. Global Warming Potential-100
Contribution to thet net impact (%)
100
80
60
40
20
0
-20
-40
-60
-80
Spring barley-straw
Willow
Alfalfa
Field emissions (N_2O)
SOC change-100 y
Seeds/Cuttings production
Field preparation
Agro-chemicals production
Harvesting and loading
Transport
Figure 3.b. Eutrophication Potential
Contribution to thet net impact (%)
100
80
60
40
20
0
Spring barley-straw
Willow
Alfalfa
Field emission (N and P related)
Seeds/Cuttings production
Field preparation
Agro-chemicals production
Harvesting and loading
Transport
Figure 3.c. NRE use, fossil
Contribution to thet net impact (%)
100
80
60
40
20
0
Spring barley-straw
Willow
Alfalfa
Seeds/Cuttings production
Field preparation
Agro-chemicals production
Harvesting and loading
Transport

Fig. 3.