

i. Time dependent climate impact of a bioenergy system - methodology development and application to Swedish conditions

ii. Time dependent climate impact of bioenergy

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Abstract

The area of dedicated energy crops is expected to increase in Sweden. This will result in direct land use changes, which may affect the carbon stocks in soil and biomass, as well as yield levels and the use of inputs. Carbon dioxide (CO₂) fluxes of biomass are often not considered when calculating the climate impact in life cycle assessments (LCA) assuming that the CO₂ released at combustion has recently been captured by the biomass in question. With the extended time lag between capture and release of CO₂ inherent to many perennial bioenergy systems, the relation between carbon neutrality and climate neutrality may be questioned.

In this paper, previously published methodologies and models are combined in a methodological framework that can assist LCA practitioners in interpreting the time dependent climate impact of a bioenergy system. The treatment of carbon differs from conventional LCA practice in that no distinction is made between fossil and biogenic carbon. A time dependent indicator is used to enable a representation of the climate impact that is not dependent on the choice of a specific characterization time horizon or time of evaluation and that does not use characterization factors, such as GWP and GTP. The indicator used to aid in the interpretation phase of this paper is global mean surface temperature change ($\Delta T_s(n)$).

A theoretical system producing willow for district heating was used to study land use change effects depending on previous land use and variations in the standing biomass carbon stocks. When replacing annual crops with willow this system presented a cooling contribution to $\Delta T_s(n)$. However, the first years after establishing the willow plantation it presented a warming contribution to $\Delta T_s(n)$. This behavior was due mainly to the soil organic carbon variation. A rapid initial increase in standing biomass counteracted the initial soil organic carbon loss.

Introduction

It is commonly accepted that altered concentrations of greenhouse gases (GHG) in the atmosphere affect the Earth's climate system. The use of fossil fuel is a main contributor to the increase in GHG concentrations in the atmosphere. In the context of climate mitigation strategies, bioenergy is considered an alternative to fossil fuels.

Bioenergy supply in Sweden has increased from 67 TWh in 1990 to 141 TWh in 2010 (Swedish Energy Agency 2011a), providing 22% of total energy input. This trend is expected to continue. The majority of the biomass used has its origins in Swedish forest (Swedish Energy Agency 2011b). Higher energy prices and increased competition for forest products are likely to increase the market share of dedicated energy crops.

Short rotation coppice (SRC) willow is a dedicated energy crop grown in Sweden as feedstock for district heating (DH) and combined heat and power plants (CHP). Willow is suitable for this purpose because it is fast growing, high yielding, has high nutrient use efficiency and comprises a workload that is compatible with other farm level activities (Mola-Yudego & González-Olabarria 2010). Other reasons for introducing willow are farm diversification, adding new biotopes to the agricultural landscape and phytoremediation. Breeding and research on willow have been actively pursued in Sweden since the 1980s. Today there are a number of high-yielding clones available on the market that are adapted to Swedish conditions.

There were about 11 000 ha of SRC willow in Sweden in 2011. An increase in the total area of SRC willow of between 100 000 to 400 000 ha in 2030 has been projected, considering the use of recently abandoned productive land as well as conversion from annual cropping systems and pasture land to dedicated energy crops (SJV 2009).

Expanding the area of dedicated energy crops will lead to a direct land use change (dLUC) and may also result in indirect land use changes (iLUC) due to market mechanisms (Njakou Djomo & Ceulemans 2012). Establishing dedicated energy crops on previous farmland is likely to affect the carbon (C) stocks in soil and biomass (Lal 2004) as well as yield levels and the use of inputs. Soil organic carbon (SOC) stocks reach a quasi-steady state after extended periods of similar land use. Two important variables controlling the steady state SOC content are the amount of input to the soil from growing biomass and the rate at which it decomposes (Paustian

et al. 1998). All of these effects can lead to climate impacts and should therefore be considered when performing a life cycle assessment (LCA) of bioenergy systems (Cherubini *et al.* 2009).

Climate impact is one of the most commonly used impact categories in the life cycle impact assessment (LCIA) phase of LCA (Cherubini & Strømman 2011), with the global warming potential using a 100 year time horizon (GWP₁₀₀) being the most commonly used characterization factor. The potential climate impact calculated in the LCIA phase is based on the life cycle inventory analysis (LCI). When performing an LCI, biogenic carbon dioxide (CO₂) is normally not considered. It is assumed that the CO₂ released has recently been captured by the biomass being used, thus closing the C cycle. However, with the extended time lag between capture and release of CO₂ inherent to many perennial bioenergy systems, the relationship between carbon neutrality and climate neutrality may be questioned (Brandão *et al.* 2012).

One issue related to time is the relative weight being given to different GHG and short lived climate forcers (SLCF) when choosing a characterization time horizon (TH), or time of evaluation (TE), to be used with a climate metric to derive characterization factors (IPCC 1991; Manne & Richels 2001; Fearnside 2002; Fuglestvedt *et al.* 2003; Shine *et al.* 2007). Absolute metrics express climate impact equivalence directly in a common physical property, such as the effect of a pulse emission of a GHG on the radiative forcing (RF) or global mean surface temperature change (ΔT_s). It can be expressed as the instantaneous value for any given point in time (TE) or integrated over a specific TH (Peters *et al.* 2011a). An important difference between an integrated and an instantaneous metric is that the integrated metric “remembers” impacts taking place throughout the entire TH while instantaneous metrics only express the climate impact at the TE. All metrics can be normalized in order to express climate impact equivalence in a reference substance, as is done when expressing GWP (Forster *et al.* 2007) or global temperature potentials (GTP, Shine *et al.* 2007) in CO₂-equivalents. Which metric to use depends on its intended application (Tanaka *et al.* 2010) and the choice of a TH or TE is ultimately subjective and can not be based solely on natural science (Fuglestvedt *et al.* 2010).

A related issue, that connects directly to how GWP is being used in LCA, is the time preference weighting of emissions. It has been recognized both in connection to LCA and carbon accounting that emissions taking place at different points in time, as well as temporary car-

bon storage and avoided emissions, will affect the magnitude of climate impacts depending on when the event takes place in relation to a specific TE (Fearnside *et al.* 2000; O'Hare *et al.* 2009; Peters *et al.* 2011a; Brandão *et al.* 2012). Standard LCA practice is to express the net value of emissions that occur at different points in time as a single score climate impact value. Characterization factors are used to convert net emissions of different GHG to a common unitless indicator value, CO₂-equivalents. This is equivalent to accounting all emissions occurring throughout the study period as if occurring in the same year (Peters *et al.* 2011a).

Several approaches have been proposed in order to account for the timing of emissions and temporary carbon storage, both in the context of carbon accounting and LCA (e.g. Moura Costa & Wilson 2000; Fearnside *et al.* 2000; O'Hare *et al.* 2009; ?; Levasseur *et al.* 2010; Cherubini *et al.* 2011; Kendall 2012). The use of dynamic characterization factors (DCF, Levasseur *et al.* 2010) and time-adjusted warming potentials (TAWP, Kendall 2012) give different weight to emissions based on the timing of emission by harmonizing the TH of each emission with the TE. These approaches are similar to the Lash-of method (Fearnside *et al.* 2000) and the fuel warming potential (FWP) proposed by O'Hare *et al.* (2009). They all require the use of a time distributed LCI for the calculation of the climate impact category indicator. The GWP_{bio} characterization factors (Cherubini *et al.* 2011), on the other hand, aims directly at quantifying the climate impact of biogenic CO₂ emissions. It combines the biomass regrowth curve with the atmospheric decay curve of a CO₂ emission. They are thus dependent on the regrowth rate and rotation length of the biomass being investigated. The concept has been expanded using probability distributions to calculate characterization factors for temporary carbon storage in wood products (Cherubini *et al.* 2012).

All the above methods use characterization factors in order to convert different GHG fluxes taking place throughout the life cycle of the study into a climate impact value, as is common practice in the LCIA phase of an LCA. Peters *et al.* (2011a) argues for the use of absolute metrics together with time distributed life cycle inventories since they can provide additional information and add valuable insights to impact assessments as well as increase transparency of an LCA. Given that the value of the climate impact category indicator results may vary for the same emission scenario depending on which characterization factor is used (Levasseur *et al.*

2012) a complementary approach to the convention of using characterization factors would be desirable in order to better understand the climate effects due to timing and type of emission when interpreting an LCA.

The aim of the present study was to assess the energy efficiency and the time dependent climate impact of a SRC willow-DH system. The objective was to represent the potential climate impact as a function of time, using an absolute and instantaneous indicator to weight climate impact between different GHG, thus avoiding the use of characterization factors and partly the choice of a specific TH or TE. This was accomplished by basing the LCIA calculations on time distributed flows including all sources and sinks within the system. Furthermore the LCI was performed in such a manner that LCIA results can also be calculated and reported in a format compliant with standard ISO 14040:2006 practice.

The potential climate impact calculated through the presented characterization method can be used as a support in the interpretation phase of the LCA study, thus complementing the use of characterization factors.

Method

System boundaries and functional unit

The study took the form of a partial LCA focusing on the energy efficiency and time dependent climate impact of a bioenergy system using SRC willow for the production of heat at a DH plant. The system boundaries included production of inputs, cultivation and harvesting of willow, biomass transportation to the DH plant and combustion of the willow chips. Activities and losses taking place after the heat produced had been delivered to a local DH distribution system were outside the system boundaries. No technical improvements or yield increases due to breeding or changed management practices were considered. The time dependent climate impact was calculated based on the three major GHG contributing to global warming: carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O), using a temporal resolution of one year. SLCF and changes in albedo, as well as indirect land use change (iLUC) effects, were excluded. The functional unit used in the impact assessment was 1 GJ of heat delivered to the local DH distribution system. The time distributed LCI and time dependent climate impact results were presented per hectare since the area unit is constant, while the energy output of the system varies between years.

System description

The system studied covered four subsequent SRC rotations, cultivated for a period of 100 years. The willow was assumed to have a coppicing cycle of three years. The time frame of the study was set to 101 years in order to include the effects of initial changes in SOC and emissions related to combustion of the final harvest. Annual net emissions of CO₂, CH₄ and N₂O from the SRC system were calculated. All upstream emissions were accounted for the year in which the activity that gave rise to them occurred.

The soil was assumed to be prepared for cultivation in autumn every 25 years, starting in year 0. Willow was planted in the following spring. All physical activities relating to the production of willow chips, collectively referred to here as operations, included soil preparation, mechanical weed control, planting, application of pesticides, fertilization and harvesting, as

well as cutting up the willow roots and stools in spring after the last harvest of each rotation. The soil was then prepared for a new rotation, which was planted the following spring. The procedures used in this study were those recommended by Gustafsson *et al.* (2006), modified according to the recommendation of Verwijst *et al.* (2010) to exclude the common practice of cutting down the plants after the establishment year to promote development of more stems. The harvesting chain consisted of direct chipping and temporary container storage at the side of the field. The containers were assumed to be transported by truck 35 km to a DH-plant. The truck had an empty return trip back to the field. All the above activities were included in operations. The input data used are summarized in Table S1.

The assumed mineral nitrogen (N) dose was based on recommendations by Aronsson & Rosenqvist (2011) and N fertilizers were not applied during the first cutting cycle. Phosphorus (P) and potassium (K) were assumed to be added according to Börjesson (2006). Fertilizer application rate and level can be found in Table S2. Chemical weed control was also assumed to be performed according to Nilsson & Bernesson (2008).

The CO₂ emitted on burning the willow chips at the DH plant was calculated based on the C stored in the live biomass. No other emissions from combustion were included. The DH-plant had an assumed efficiency of 85%. The willow chips were combusted within a few months upon delivery. Dry matter losses were assumed to be 3 % based on an average storage period of 60 days (Elinder *et al.* 1995).

The potential climate impact of a reference case was also calculated. In this case the same amount of energy was produced using coal. The district heating plant was assumed to have an energy efficiency of 85% and the GHG emissions were taken from the recommendations in Uppenberg *et al.* (2001).

Previous land use

Two different scenarios were compared in order to assess the influence of previous land use on the time dependent climate impact. The same management methods, yields and inter-annual distribution of emissions were assumed in both scenarios.

Scenario 1. Annual crops. In this scenario the field was assumed to have been cultivated with

an annual crop rotation for an extended period of time, such that SOC had reached steady-state. Equal proportions of spring and winter cereals were used as a proxy for the annual crop rotation to derive input values giving the steady state values.

Scenario 2. 20-year-old fallow. Between 1990 and 2006, approximately 185 000 ha of agricultural land were put into fallow in Sweden (SJV 2008). In this scenario the field was assumed to have been put into fallow 20 years in the past. Before being put into fallow the same conditions as for scenario 1 were assumed. This means that SOC was not in steady state.

Life cycle inventory analysis

To determine the effect of timing of emissions as well as type of emission additional information is required when performing the LCI than what is normally required in a standard LCA. We followed the same basic methodology as in Fearnside *et al.* (2000); O'Hare *et al.* (2009); Levasseur *et al.* (2010) and Kendall (2012), developing a time distributed LCI. The annual net flux of each GHG was determined for all sources and sinks and recorded as emission impulses (EI). The annual net flux to the atmosphere of GHG x in the i^{th} year of the study is referred to as EI_{x_i} [kg].

Carbon fluxes in soil and standing biomass

Carbon fluxes due to variations in standing biomass were calculated based on expected harvest, inter-annual growth rate and C allocation patterns derived from lysimeter experiments (Rytter 2001). An expected harvest of 20 Mg DM · ha⁻¹ for the first coppicing cycle and 30 Mg DM · ha⁻¹ for subsequent coppicing cycles was assumed (Nilsson & Bernesson 2008). Carbon allocation data and the share of the expected harvest attributed to each year of the coppicing cycle can be found in Table S3.

The standing biomass was divided into the following pools: stems, leaves, coarse roots and stumps. The wood chips were assumed to be burnt in the year of harvest, returning all C in the stem pool to the atmosphere.

Carbon fluxes due to SOC changes were calculated using the ICBMr dynamic soil carbon model (Andr  n *et al.* 2004), adjusted for a SRC willow system. All biogenic C fluxes to and from the atmosphere were assumed to be in the form of CO₂.

ICBMr is a version of the introductory carbon balance model (ICBM) (Andr  n & K  tterer 1997) that has been adapted for use with variable annual input. The ICBMr model consists of two C containing pools: young, Y , and old, O . The differential equations governing the development of these can be found in the supporting information (equations S1 & S2). Carbon enters the Y -pool as input, i , mainly from litter, dead roots and root exudates. The amount of C leaving Y and O is determined by two decay constant, k_Y and k_O . These were originally calibrated using data from the Ultuna long-term frame trial (Kirchmann *et al.* 1994). Decay rates of Y and O are also determined by a third variable, r_e , which represents external factors, such as climate. The fraction of the flow leaving Y that enters O is determined by a parameter, h , which represents the decomposability of i .

According to various studies belowground inputs contribute more to refractory SOC than aboveground residues (Johnson *et al.* 2006; K  tterer *et al.* 2011). Consequently, the model was adapted by using two parallel Y -pools, aboveground (suffix a) and belowground (suffix b), receiving separate inputs (i_a and i_b) associated with different h values (h_a and h_b). In the model $h_b = 2.3 \times h_a$ (K  tterer *et al.* 2011). Equations 1 and 2 are used to calculate the SOC stock with a yearly time step:

$$Y_{[a,b]}(t) = (Y_{[a,b]}_{t-1} + i_{[a,b]}_{t-1}) \cdot \exp^{-k_y r_e} \quad (1)$$

$$O(t) = \left(O_{t-1} - \left(\frac{h_a \cdot k_y}{(k_o - k_y)} \cdot (Y_{a_{t-1}} + i_{a_{t-1}}) + \frac{h_b \cdot k_y}{k_o - k_y} \cdot (Y_{b_{t-1}} + i_{b_{t-1}}) \right) \right) \cdot \exp^{-k_o r_e} \\ + \left(\frac{h_a \cdot k_y}{k_o - k_y} \cdot (Y_{a_{t-1}} + i_{a_{t-1}}) + \frac{h_b \cdot k_y}{k_o - k_y} \cdot (Y_{b_{t-1}} + i_{b_{t-1}}) \right) \cdot \exp^{-k_y r_e} \quad (2)$$

The i_a fraction comprises leaf litter, while i_b comprises fine root turnover and the accumulated coarse roots and stumps when the plantation is broken up after each rotation. The fine root turnover rate was reported by Rytter & Rytter (1998) to be between 4.9-5.8 yr⁻¹. The standing

biomass pool, coarse roots and stumps, was assumed to accumulate C during the first cutting cycle of every rotation and remain at a constant level thereafter. At the end of a rotation (every 25 years), this C was transferred to ICBMr. The decomposability of willow litter was assumed to be similar to that found in litter bag experiments performed on willow leaves (Šlapokas & Granhall 1991a,b). $r_e = 1$ was kept constant in all simulations and reflects a location in the Mrdalen region of Sweden.

The input used to establish initial SOC content is based on previous land use and is calculated using the allometric function $i_a = a + s \cdot H$ (Andr  n *et al.* 2004) for straw and residues, where a and s are crop-specific parameters and H is the C content of the harvested crop (Table S4). Belowground input, i_b , is calculated using equation 3 (K  tterer *et al.* 2011):

$$i_b = \frac{H \times R_m \times R_{RE}}{1 - R_{RE}/0.85} \quad (3)$$

where R_{RE} is the relative C fraction allocated to roots, including rhizodeposition, and R_m is the root mass fraction accounted for. The value of R_{RE} is crop-specific and was taken here from K  tterer *et al.* (2011). The C input along the entire soil profile is included by setting $R_m = 1$. A C content of 50% of DM is used for all crops.

In scenario 1, the total C input giving the initial SOC content level was approximately 4.0 Mg C · ha⁻¹ · yr⁻¹ and in scenario 2 it was approximately 1.8 Mg C · ha⁻¹ · yr⁻¹. The initial SOC levels can be found in Table S5.

Since altering the relative contribution of aboveground and belowground input to the O -pool affects total SOC stocks, the k -values were recalibrated using data from the Ultuna long-term frame trial. Before recalibration, the SOC content of the top soil was converted to the 1956 equivalent top soil depth based on changes in soil density (K  tterer *et al.* 2011), assuming a linear change in soil density between 1956 and 2009. Annual harvest data between 1956 and 2010 were used in the calibration (T. Kerer, personal communication 2012). r_e was set to 1 for all years and treatments except for the bare fallow where it was set to 1.1. The annual below ground input in the bare fallow was set to 40 kg C · ha⁻¹ · yr⁻¹ when optimizing k_O . The root mass between 0 and 20 cm depth (R_m in equation 3) was set to 0.71 for all plant species (K  tterer *et al.* 2011).

N₂O emissions

A surplus of inorganic N in the soil causes N₂O emissions. Both direct and indirect emissions result from N fertilizer application. Direct emissions occur directly in the field due to fertilization as well as from decomposition of aboveground and belowground crop residues. Indirect emissions are generated from leached N and from ammonium that volatilizes and later redeposits (IPCC, 2006). To include the emissions caused by fertilization in the SRC system the IPCC (2006) default values were used. These state that 1% of the N applied as mineral fertilizer or of the N content in residues is converted to N₂O, and 30% of the applied N is leached. Of this fraction, 0.75% is converted to N₂O. The fraction of applied N that volatilize as ammonium was adjusted to reflect Swedish conditions, using a value of 1.2% (Ahlgren *et al.* 2011). For calculation of N₂O emissions from organic matter decomposition, a N-content of 2.5% in litter and 0.43% in stems was assumed (Weih & Nordh 2005). Fine roots were assumed to have the same N-content as stems.

Life cycle impact assessment

To fulfill the goal and scope of this study a time dependent, absolute and instantaneous indicator was used, in addition to characterization factor based GWP. The global mean surface temperature change (ΔT_s) was chosen as a climate impact category indicator for this purpose. ΔT_s is found one step further down the cause and effect chain from emission to climate impact than RF. Consequently, additional modeling and assumptions are embedded in using it as an indicator. This increases uncertainty, but at the same time adds additional value in terms of increased policy relevance. Temperature is also easier to relate to than RF for most people, which can be of value for communication purposes. The temperature response differ from the RF in a fundamental way, which is important when interpreting the climate impact category indicator. The later only includes the energy entering the climate system in relation to a specific point in time in the past, while the former also includes the thermal inertia of the climate system as well as the energy leaving the climate system as a new equilibrium approaches (Peters *et al.* 2011b).

Energy efficiency

The energy efficiency of the system was calculated as the energy delivered to the DH distribution system relative to the primary energy input to the system. This is equivalent to the energy ratio definition found in Djomo *et al.* (2011). Energy input in the production of the feedstock, transportation to the DH plant, as well as storage and energy conversion losses were included. The higher heating value of the feedstock was used in this study.

Global Warming Potential

The GWP₁₀₀ characterization factors in the IPCC AR4 (2007) were applied to the net emissions of each GHG. These were summed up to calculate the GWP value for each scenario, as well as the reference case. These values are equivalent to those calculated when performing a standard LCA.

Global mean surface temperature change

The characterization model used in this study to convert GHG emissions to ΔT_s is based on the models used in Boucher & Reddy (2008); Boucher *et al.* (2009) and Fuglestedt *et al.* (2010). In the following $\Delta T_s(n)$ is used as abbreviation for the time dependent global mean surface temperature change due to a specific emission scenario used in a specific case study, where n is the year relative to the first year of the study time frame. Three steps were followed to determine $\Delta T_s(n)$, in addition to the time distributed LCI previously described:

In step one, the change in the atmospheric concentration of a GHG, $f_{x_i}(t)$, due to EI_{x_i} at $t = 0$ was calculated as:

$$f_{x_i}(t) = EI_{x_i} \cdot f_x(t) \quad [\text{kg}] \quad (4)$$

The impulse response function (IRF) ($f_x(t)$) in equation 4 describes the change in GHG concentration due to a unit impulse of a gas at $t=0$ (Ramaswamy *et al.* 2001). For CO₂, it is a compound function (equation 5) with three exponential decay terms ($j = [1, 2, 3]$), each with a specific atmospheric decay constant, τ_j . The weight of each term is given by a_j . A fourth term,

a_0 , gives the residual fraction of an impulse that is not removed from the atmosphere:

$$f_x(t) = a_0 + \sum_{j=1}^k a_j \cdot \exp^{-\frac{t}{\tau_j}} \quad [\text{unitless}] \quad (5)$$

The same parameter values as in Forster *et al.* (2007), based on the “Bern” carbon cycle model, were used here (Table S6). N_2O and CH_4 were modeled using a simple exponential decay model, with $\tau = 114$ years for N_2O and $\tau = 12$ years for CH_4 .

The CO_2 -indirect effect of CH_4 was included in the model by adding the fraction of previously emitted CH_4 that is broken down between the years $i - 1$ and i to $EI_{\text{CO}_2_i}$. All emitted CH_4 was assumed to be oxidized into CO_2 .

In step two, the change in radiative forcing, $RF_{x_i}(t)$, was calculated based on the change in the atmospheric GHG concentration. This was done by multiplying the radiative efficiency, $RE_x [\text{W} \cdot \text{m}^{-2} \cdot \text{kg}^{-1}]$, of a gas by $f_{x_i}(t)$:

$$RF_{x_i}(t) = RE_x \cdot f_{x_i}(t) \quad [\text{W} \cdot \text{m}^{-2}] \quad (6)$$

RE_x was calculated using the simplified expressions given in Ramaswamy *et al.* (2001). Constant background concentrations were assumed using the values given in the IPCC AR4 (Forster *et al.* 2007).

In step three, the global mean surface temperature change in the n^{th} year of the study, $\Delta T_s(n)$, was calculated as the sum of all individual temperature response functions due to previous EI_{x_i} , $\Delta T_s^{x_i}(t)$ with $i \leq n$:

$$\Delta T_s(n) = \sum_{x=1}^3 \sum_{i=1}^n \Delta T_s^{x_i}(t) \quad [\text{K}] \quad (7)$$

The $\Delta T_s^{x_i}(t)$ term in equation 7 represents the characteristic temperature response to an EI_{x_i} of GHG x in the year i . This is the result of the $RF_{x_i}(t)$ and the climate system temperature response to a perturbation of the RF . The climate system temperature response to a perturbation of the RF is represented by a temperature response function, $\delta T_s(t)$, which describes the temperature response to a unit increase in the RF . It is possible to find $\Delta T_s^{x_i}$ by performing a convolution between $RF_{x_i}(t)$ and $\delta T_s(t)$, assuming a linear and time-invariant (LTI) system:

$$\Delta T_s^{x_i}(t) = \int_{t-\tau}^t RF_{x_i}(\tau) \delta T_s(t-\tau) d\tau \quad [\text{K}] \quad (8)$$

In this study the $\delta T_s(t)$ IRF presented in Boucher & Reddy (2008) was used to represent the climate system response to a perturbation of the radiative balance. Equation 9 was calibrated by Boucher & Reddy against the Had3CM climate model. It has two climate response time scales ($d_1 = 8.4$, $d_2 = 409.5$) and a total equilibrium climate sensitivity of $1.06 \text{ K} \cdot (\text{W} \cdot \text{m}^{-2})^{-1}$, which is given by the sum of the c_j coefficients ($c_1 = 0.631$, $c_2 = 0.429$):

$$\delta T_s(t) = \sum_{j=1}^2 c_j/d_j \cdot \exp\left(-\frac{t}{d_j}\right) \quad (9)$$

Equation 8 was solved using the built-in Laplace transform functions in the maxima software (Maxima 2011), giving:

$$\begin{cases} \Delta T_s^{CO_2i}(t) = EI_{CO_2i} \cdot RE_{CO_2} \cdot \left(k_0 + \sum_{j=1}^5 k_j \exp\left(-\frac{t}{\tau_j^{CO_2}}\right) \right) & [\text{K}] \\ \Delta T_s^{CH_4i}(t) = EI_{CH_4i} \cdot RE_{CH_4} \cdot \left(\sum_{j=1}^3 m_j \exp\left(-\frac{t}{\tau_j^{CH_4}}\right) \right) & [\text{K}] \\ \Delta T_s^{N_2O_i}(t) = EI_{N_2O_i} \cdot RE_{N_2O} \cdot \left(\sum_{j=1}^3 n_j \exp\left(-\frac{t}{\tau_j^{N_2O}}\right) \right) & [\text{K}] \end{cases} \quad (10)$$

The parameter values in Table 1 were used to calculate $\Delta T_s^{x_i}(t)$. The resulting $\Delta T_s^{x_i}(t)$ is shown in Fig. 1 for EI_{x_i} of all three GHG exerting an effect of $1 \text{ W} \cdot \text{m}^{-2}$ on the radiative balance at $t = 0$.

Results

Life cycle inventory analysis

There were net emissions of -8.7 and -9.8 kg CO₂ per GJ heat delivered to the DH distribution system in scenario 1 and 2, respectively, indicating that the system was a net sink of carbon in both scenarios. The contribution from field operations to the net emissions of CO₂ was 3.1 kg in both scenarios. Net emissions of CH₄ and N₂O were also identical in both scenarios and amounted to 2.3×10^{-3} and 1.2×10^{-2} kg, respectively.

The reference case caused net emissions of 111 kg CO₂, 1.29 kg CH₄ and 1.4×10^{-2} kg N₂O per GJ heat delivered.

The GHG flux profile in the time distributed LCI showed cyclic repetitions due to the three-year coppicing cycle and 25-year rotation period (Figs. 2 and 3). The initial three-year period had a different profile, since most of crop management operations take place during the establishment phase of a willow rotation.

GHG fluxes from subsequent rotations were identical for all parts of the system, except for soil CO₂ fluxes. This means that the period 1-25 years in Fig. 2 can also be read as 26-50, 51-75 and 76-100 years.

There was a net decrease in the SOC stock during the first years of the plantation. This led to an initial flux of CO₂ from the soil to the atmosphere and can be explained by the conversion from an annual cropping system to the SRC willow system. The former leaves a large pool of easily decomposed SOC while the later contributes with little C input during the establishment years. Apart from this initial release of CO₂, the rate of C sequestration was higher in earlier rotations (Fig. 3). Following each harvest there was a net flux of CO₂ from the soil to the atmosphere, when input was smaller than decay. This effect was larger directly after ending a rotation, since growth rate, and thus the C input, was lower during the first cutting cycle.

Energy efficiency

Total energy content of the harvested willow produced in one rotation was 4593 GJ (assuming HHV = 19.96 GJ·Gg⁻¹ DM). A total amount of 156 GJ of energy was put into the system during

one rotation. This input included all energy used in upstream processes and production of the willow as well as transportation to the DH plant. Including the storage and energy conversion losses, this gives a total energy efficiency of 24.3 in terms of energy delivered from the SRC willow-DH system related to the energy inputs.

Global Warming Potential

The GWP values, expressed per GJ of heat delivered to the DH distribution system, were -5.01 kg CO₂-eq for scenario 1, where the willow plantation replaced an annual cropping system and -6.01 kg CO₂-eq for scenario 2, where the willow plantation replaced a 20 year old fallow. For the reference case, where the heat was produced in a conventional coal fired DH plant, the GWP value was 148 kg CO₂-eq per GJ of heat delivered.

Global mean surface temperature change, $\Delta T_s(n)$

The $\Delta T_s(n)$ for the different parts of the system are shown as individual temperature response curves, $\Delta T_s^x(n)$, in Fig. 4. The absolute contribution from the $\Delta T_s^{CH_4}(n)$ was very small in the SRC willow-DH system (Fig. 5).

The CO₂ and N₂O emissions from operations represented fossil fuel inputs to the system. These emissions had a continuous impact on $\Delta T_s(n)$ throughout all four rotations, representing a warming contribution to the global mean surface temperature (Fig. 4). In contrast, the accumulation of C in the standing biomass pool during the first rotation had a cooling impact on $\Delta T_s(n)$ (Fig. 4). This effect rapidly decreased after the first rotation, which can be explained by the shape of the temperature response to a pulse emission of CO₂. Figure 1 shows that there is a peak in the temperature response within 20 years from the time of a pulse emission. Emissions that occurred so far back that they have already passed their peak at the TE in a specific emission scenario actually have a reverse impact on the rate of change of $\Delta T_s(n)$, relative to their initial impact.

The response from the increasing SOC levels dominated in the system, dictating the trend in $\Delta T_s(n)$ (Fig. 6). An initial decrease in the SOC stock (Fig. 3) caused a net positive total $\Delta T_s(n)$ in the first years after conversion to willow cultivation (Fig. 6). However, the carbon debt was

quickly repaid through the increased C input to the soil from the willow. This was manifested in a net negative total $\Delta T_s(n)$ after a few years (Figs. 4 and 6). The increased C stock in the standing biomass counteracted the SOC loss and made the time where the total $\Delta T_s(n)$ turns net negative occur earlier than if this effect would not have been included.

The SRC willow-DH system represented a cooling contribution to the global mean surface temperature throughout most of the time frame of the study. Scenario 2 had a larger impact on $\Delta T_s(n)$ than scenario 1 (Fig. 6). The entire difference between the two scenarios was due to the $\Delta T_s^{CO_2}(n)$ of SOC (Fig. 4).

Discussion

The results from the present study show that the establishment of a SRC willow plantation on a typical mid-Swedish clay soil to produce feedstock for a DH plant will have a cooling influence on the global mean surface temperature over the time frame of the study. This effect is due to the increase in C stocks of the live biomass and the long term SOC increase. Both the GWP values and ΔT_s indicate that replacing a 20-year old fallow has a larger climate impact than replacing annual crops under the assumptions used in this study. This is due to the soil of the fallow scenario having a lower initial SOC level than that under annual crops.

The energy efficiency of the SRC willow-DH system is 24 times the energy input, which places it right in the middle of the 21 studies of SRC poplar and willow studies compared by Djomo *et al.* (2011).

The inability of GWP to capture the effects of timing of emissions on the climate impact at a specific point in time, when used in an LCA context, is directly related to the use of a time horizon having a fixed length, giving equal weight to all emissions irregardless of when they occur throughout the time frame of the study. The use of a time distributed LCI, together with time dependent characterization factors, such as DCF and TAWP, resolves this issue to some extent since they give different weight to emissions based on when they occur in relation to a specific moment in time (TE). They are able to express biogenic CO₂ fluxes within a given bioenergy system in a single score climate impact value since these CO₂ fluxes have a typical time distributed pattern. The potential climate impact is however reduced to a single value which gives no further information about the shape of the time dependent climate impact. When combining the time distributed LCI with a time dependent climate impact indicator, as was done in this study, additional insight is obtained about the behavior of the system and how the relative importance of system components change over time (Fig. 4). This also removes some of the subjectivity included in the characterization factors. The explicit representation of the potential climate impact as a function of time partly removes the responsibility of the practitioner to choose TH or TE, leaving this choice to the interpreter of the results.

The absolute and instantaneous climate impact indicator $\Delta T_s(n)$ used in this study to aid in the interpretation of the results uses the temperature change due to a point emission of a GHG

to express the results. There is no further need for weighting of different GHG, which simplifies the inclusion of other climate forcers, such as SLCF and albedo, if desired (Fuglestvedt *et al.* 2010). Furthermore, less ambiguity is left as to which climate response is being addressed (Fuglestvedt *et al.* 2003).

The methodology followed to calculate $\Delta T_s(n)$ was chosen since it reduces the complexity of a global climate model (GCM) to a computational size that is manageable within a bioenergy LCA context. We followed the same methodology as in Boucher & Reddy (2008), using an IRF to calculate ΔT_s , since this can fully emulate any emission impulse response of a more complex GCM (Hansen *et al.* 2011). This gives flexibility to the LCA in that it does not imply the use of ΔT_s . It could just as well express climate impact equivalence in another indicator, such as change in sea-level, cloudiness or precipitation pattern (Hooss *et al.* 2001; Joos *et al.* 2001). It would be accomplished by switching the IRF in the characterization model to represent another response, also derived from a GCM.

The rapid assessment of many alternative scenarios is desirable when performing an LCA. IRFs fulfill this requirement. However, IRFs restricts the interpretation of the results, since a linear and time-invariant system is assumed. Therefore, the results should not be interpreted as an absolute contribution to ΔT_s in the future. Rather, they represent how the system would affect the climate if no other variables were to change in the climate system. As an example, the shape of the $\Delta T_s(n)$ curve in Fig. 6 would be affected by an increased background concentration of CO₂, both as a consequence of altered uptake rates by different sinks (Prentice *et al.* 2001) and the altered radiative efficiency of CO₂ (Ramaswamy *et al.* 2001).

In the present study the temperature response was modeled as in Boucher *et al.* (2009), using the same IRF (equation 9) for all three gases. It is important to remember that the climate sensitivity, representing the equilibrium temperature response to a change in RF, might vary between different climate forcers. This could be incorporated in the calculation of $\Delta T_s(n)$ by multiplying equation 10 with the specific climate efficacy value of each climate forcer. The efficacy is defined as the ratio of the climate sensitivity for a specific climate forcer to that of CO₂ (IPCC 2007). In this study it was not included since the climate efficacy values of different climate forcers is currently not consistent between different GCMs (Fuglestvedt *et al.*

2010).

The methodology followed in this paper keeps track of both the timing and climate response of each emission impulse. All sinks and sources of the studied system are included in the LCI models used. The distinction between fossil and biogenic C is not necessary in the time distributed LCI when it is combined with a time dependent characterization model, such as that used to calculate $\Delta T_s(n)$. This is consistent with the fact that, once in the air, a GHG will affect the climate, regardless of its origin. This also enables the CO₂-indirect effect of CH₄ (Boucher *et al.* 2009) to be included by simply returning it as input to the C model. Although there are very few CH₄ emissions in the SRC willow-DH system studied here, it can be seen in Fig. 6 that the CO₂-indirect effect of CH₄ is highly time dependent and increases over time (Boucher *et al.* 2009). This is due to the CH₄ being gradually broken down, as well as to the different shapes of the IRFs of the two gases (Fig. 1). The IRF of CO₂ (equation 5) has a residual fraction that is not removed from the atmosphere, while methane is broken down completely.

The initial SOC levels calculated (Table S5) were similar to those presented by Hillier *et al.* (2009) and Kimming *et al.* (2011). Soil organic carbon accumulation rate was lower than that found in Grogan & Matthews (2002), while the SOC level after 100 years was higher. This can be explained by the higher initial SOC content in the SRC willow-DH study. Hillier *et al.* (2009) and Grogan & Matthews (2002) both used other SOC models in their studies. The SOC model used here (ICBMr) could easily be replaced by another soil carbon model within the methodological framework of this study.

Soil is not an infinite carbon sink. Eventually a new SOC steady state will be reached (Lal 2004). What is not shown here is that the rate of growth of $\Delta T_s(n)$ will turn positive after the fourth rotation as an effect of the system being essentially fossil fuel-driven. The rate of growth of $\Delta T_s^{N_2O}(n)$ and $\Delta T_s^{CO_2}(n)$ from operations are such that these will always represent a warming contribution to global mean surface temperature. However, the rate of growth of $\Delta T_s^{CO_2}(n)$, which represent a cooling contribution to the global mean surface temperature, will decrease when the SOC approaches a new steady state. In the case study, both scenarios had identical steady states. If reverting back to previous land use, all sequestered C would eventually be returned to the atmosphere.

In the present study, the initial negative rate of growth of $\Delta T_s(n)$ due to the increased standing biomass C pool turned slightly positive after the first rotation (Fig. 4). This effect is not accounted for when using GWP to represent climate impact in an LCA, effectively interpreting biogenic carbon as climate neutral. The $\Delta T_s^{CO_2}(n)$ from standing biomass in Fig. 4 illustrates rather clearly that biogenic carbon is not climate neutral when a land use change occurs.

The results from using the methodology presented in this paper showed that it may be used to aid in the interpretation of the studied system, as was done here. The time dependent absolute and instantaneous climate impact indicator used in this paper is not an alternative to characterization factor based climate impact indicator results, but complements them and enhances the usefulness of the LCIA results. Apart from aiding in the interpretation of the results, the LCA practitioner develops a profound understanding of the system when developing the time distributed LCI. This should be beneficial for the quality of the LCI. Furthermore, the LCI could serve as input for analysis further down the emission to damage response chain, such as ecosystem responses, damage costs or cost benefit analysis. The use in LCA of a time dependent indicator directly related to a climate response could also increase the usefulness of the results, since it enables rapid assessment of the contribution from different bioenergy development scenarios in relation to specific climate goals, e.g. the EU 2°C climate target (EU 2008).

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Tables

Table 1: Parameters calculated using Maxima (2011) to be used in equation 7 to calculate ΔT_s .

	$i = 0$	$i = 1$	$i = 2$	$i = 3$	$i = 4$	$i = 5$
k	0.230	-0.020	-0.680	0.384	0.091	-0.005
m		-1.984	1.972	0.012		
n		-0.643	0.487	0.156		
τ^{CO_2}		1.186	8.4	18.51	172.9	409.5
τ^{CH_4}		8.4	12	409.5		
τ^{N_2O}		8.4	114	409.5		

Figure legends

Fig. 1 Temperature response curves of the three GHG studied due to pulse emissions at $t = 0$. The response curves were calculated with equation 10 using emission impulses of a magnitude such that the initial perturbation of the RF is $1 \text{ W} \cdot \text{m}^{-2}$ at time 0 for each GHG ($EI_{x_0} = 2.6 \text{ Pg N}_2\text{O}$; 570 Pg CO_2 ; 5.6 Pg CH_4).

Fig. 2 Emission impulses from operations for one rotation, including production and use of inputs for the preparation of the field, mechanical weed control, planting, application of pesticides, fertilization, and harvesting, as well as cutting up the roots and stools after the last harvest of each rotation. N_2O emissions are divided into biogenic (solid black) and production of inputs (white).

Fig. 3 CO_2 fluxes in scenario 1 due to carbon stock changes in the SOC and variations in standing biomass.

Fig. 4 Temperature response due to the different sinks and sources of GHG in the SRC willow-DH system.

Fig. 5 Contribution of CH_4 to $\Delta T_s(n)$, both through direct effect, based on the exponential decay model (Ramaswamy *et al.* 2001), and the indirect effect from the CO_2 resulting from the atmospheric decay of CH_4 .

Fig. 6 Change in $\Delta T_s(n)$ for both scenarios as a result of all GHG fluxes in the SRC willow-DH system.

Supporting information

Supporting.pdf contains the input data used when calculating the life cycle inventory and $\Delta T_s(n)$ as well as calculated SOC levels, the differential equations governing the ICBMr model and a list of abbreviations used within the article.