An assessment of the torrefaction of North American pine and life cycle greenhouse gas emissions

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ABSTRACT

Bioenergy is increasingly being used to meet EU objectives for renewable energy generation and reducing greenhouse gas (GHG) emissions. Problems with using biomass however include high moisture contents, lower calorific value and poor grindability when compared to fossil fuels. Torrefaction is a pre-treatment process that aims to address these issues. In this paper four torrefaction treatments of pine were performed and a mass–energy balance calculated. Using experimental data, a pellet production supply chain incorporating torrefaction was modelled and compared to an existing wood pellet system to determine life-cycle GHG emissions. Two utility fuels, wood chips and natural gas, were considered to provide process heat in addition to volatile gases released during torrefaction (torgas). Experimental results show that torrefaction reduces the moisture content and increases the calorific value of the fuels. Increasing torrefaction temperature and residence time results in lower mass and energy yields. GHG emissions reduce with increasing torrefaction severity. Emissions from drying & torrefaction and shipping are the highest GHG contributors to the supply chain. All 4 torrefaction conditions assessed outperformed traditional wood pellet supply chain emissions but more land is required which increases with temperature and residence time. Sensitivity analysis results show that emissions increase significantly where natural gas is used for utility fuel and no torgas is utilised.

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1. Introduction

Several EU countries are endeavouring to increase the use of renewable energy and reduce greenhouse gas emissions by 2020 with further targets set for 2030 [1, 2]. In addition to the targets set out in the European Renewable Energy Directive, the UK is also bound by legal framework to reduce its GHG emissions by at least 80% relative to 1990 levels by 2050 in accordance with the Climate Change Act 2008 [3]. In fulfilling these emissions reduction targets, it is widely accepted that bioenergy will play a significant role as outlined in the UK bioenergy strategy [4] and as a result is being increasingly mobilised to help achieve these policy goals domestically as well as across several EU states.

The energy sector was responsible just under a third of UK GHG emissions in 2013 [5], therefore ways in which it can be reduced must be explored. The use of solid biomass for energy has undergone rapid expansion in the past few years owing to its relatively easy and cost-effective incorporation in to existing solid fuel supply chains (e.g. coal supply chains). In addition, when compared with other sources of renewable energy, biomass is an attractive option being a carbon carrier that can be stored and brought online when required and so alleviates issues of intermittency associated with solar and wind technologies, for example. Owing to limited and competing land availability in the EU however, generous subsidies for renewable electricity and heat have led to a rapid increase in biomass imports to the UK and EU [6–8]. Despite this, there can be challenges associated with using biomass as a fuel [9] with issues including high moisture content (MC), low calorific value (CV), low bulk density, poor grindability and heterogeneity relative to fossil-based fuels which may cause problems with logistics, bulk handling and transportation [10, 11]. Densification treatments are therefore normally implemented to ‘raw’ biomass to help alleviate these problems with conventional wood pellets currently the favoured form of biomass for transportation over long distances. Ofgem reported that in 2011–12 1.3 million tonnes of wood pellets were imported to the UK under the Renewables Obligation (RO), with over 90% coming from North America [12]. Demand for wood pellets has rapidly increased over
the last decade and is projected to continue expanding due to incentives for bioenergy in the UK and EU [3,8]. Nevertheless, conventional wood pellets still retain some of the inherent problems associated with biomass such as absorption of moisture upon transportation and storage, which can cause pellets to become mouldy and disintegrate [13]. Additional treatments can therefore be considered in lieu or in combination with pelletisation. Torrefaction of biomass is one such treatment that goes some way to address these issues. Torrefaction, sometimes called ‘mild pyrolysis’ is a pre-treatment process where biomass is heated in the absence of oxygen at low heating rates (<50 °C/min) to temperatures between 200 and 300 °C to produce a darkened material with improved chemical and physical properties [14]. During torrefaction, moisture and oxygen-rich volatile materials with low calorific value are driven off resulting in a reduction in hydroxyl groups (that form hydrogen bonds with moisture) and a greater mass loss to energy loss producing a more energy dense fuel. The loss of these volatile compounds are mainly ascribed to decomposition of the hemicellulose fraction, which binds the cellulose fibrils in the cell wall providing structural integrity and so their partial or full decomposition results in a fuel with improved grindability characteristics [15–17]. It is suggested that torrefaction combined with pelletisation is thus preferable to untreated wood pellets as a result of lower moisture, higher heating value, improved bulk density and wider handling and transport benefits [9,18]. An additional benefit is the use of torrefied pellets in existing power plants. Torrefied pellets can be ground to a fine dust with relatively minor modifications in existing coal mills, whereas conventional pellets require specialised mills and higher amounts of energy for grinding [19]. A disadvantage however is the possibility of fine dust formation during handling, with associated explosion risk. Durability of pellets is therefore another important consideration.

While torrefaction improves the chemical and physical properties of raw biomass, as the process often requires drying of the fuel prior to torrefaction, the process is overall endothermic and so requires an energy input. Although most of the original energy content of the fuel is contained within the solid torrefied product, during torrefaction some energy will be lost in the volatile materials evolved which can be combusted to provide some of the heat requirements of the process. In the case where the gases produced do not meet the heat demand for drying and torrefaction, an additional source of heat is required. It must be determined therefore whether the benefits of torrefaction outweigh any potential energy penalties paid.

Utilising torrefied biomass will also have other implications along the bioenergy supply chain from the amount of feedstock required to its impact on pellet production and energy delivered to the end-user. These impacts become significant when considering life-cycle emissions and must remain within sustainability guidelines [7]. To assess the potential for torrefied wood being used as pellets, this paper combines experimental work on the torrefaction of North American pine with whole systems assessment of greenhouse gas (GHG) emissions from the supply chain. In this study, North American pine has been torrefied under 4 different conditions to determine the changes in fuel properties and a mass and energy balance for each process determined. Using these data, the energy requirements for the torrefaction processes have been modelled and incorporated in to a bioenergy supply chain in which the feedstock is harvested, torrefied and pelletised in the United States and transported to the UK to determine the life-cycle GHG emissions for electricity generation. The results are then compared with a conventional wood pellet supply chain and a sensitivity analysis performed to assess key assumptions and data uncertainties.

2. Methodology

The methodology described below is split into two main sections being the experimental work and the greenhouse gas emission assessment.

2.1. Experimental

2.1.1. Samples

The fuel used in this study was pine wood (de-barked) grown in North America and sourced from a UK power station. The sample was in the form of wood chips in the size range 5–30 mm.

2.1.2. Torrefaction of pine chips

The torrefaction experiments were conducted using a three zone electrically heated horizontal tube furnace with an internal diameter of 75 mm and 750 mm in length (Elite Thermal Systems). The heated zone, controlled by three PID controllers (Eurotherm, Schneider Electric), is approximately 575 mm in length. A full description of the experimental equipment can be found in [20]. The temperature in the furnace was monitored using three thermocouples at 20 cm intervals inside the reactor tube, measuring the temperature of both the inert gas and the sample with temperature profiles recorded for each experiment. In each torrefaction experiment, approximately 100 g of untreated sample was weighed and placed in a reactor tube 800 mm in length with an internal diameter 60 mm, with the sample positioned within the heating zone of the furnace. Each torrefaction experiment was performed under a flow of nitrogen (1.2 L/min) controlled using a valve and flow meter. The samples were heated at a rate of 10 °C/min to 150 °C and held at this temperature for one hour to dry before heating at the same rate to the final temperature where the sample was then held for the desired residence time. The torrefaction conditions performed in these experiments are shown in Table 1.

The residence time in this instance begins when the gas temperature has reached the desired final temperature and held for the desired residence time after which the reactor tube was quickly removed from the furnace and quenched with an increased flow of N2 to ensure rapid cooling. It must be noted that during some torrefaction experiments, the temperature recorded from the thermocouples was in some cases 15 °C higher than the set temperature indicating exothermic activity during torrefaction. Once cool, the torrefied sample was carefully removed from the reactor tube and weighed. The mass yields (dry basis) for each experiment were calculated using the following equation:

$$\eta_{\text{mass}} = \left( \frac{m_{\text{treated.dry}}}{m_{\text{untreated.dry}}} \right) \times 100 \quad (1)$$

where $m_{\text{treated.dry}}$ and $m_{\text{untreated.dry}}$ correspond to the mass of the treated and untreated biomass (dry basis) before and after torrefaction respectively.

| Condition name | Temperature (°C) | Residence time (min) |
|----------------|-----------------|----------------------|
| 250-30         | 250             | 30                   |
| 270-30         | 270             | 30                   |
| 270-60         | 270             | 60                   |
| 290-30         | 290             | 30                   |

Table 1: Summary of the torrefaction conditions used in torrefaction experiments.
2.1.3. Characterisation of untreated and torrefied fuels

2.1.3.1. Proximate and ultimate analysis. The moisture, volatiles and ash contents of the untreated and torrefied fuels were determined using the methods laid out in the following European Standards [21–23]. The C, H, N and S contents of untreated and torrefied fuels were measured using a CE Instruments Flash EA 1112 Series elemental analyser according to the European Standard method laid out in [24]. The oxygen contents were determined by difference.

2.1.3.2. Bomb calorimetry. The higher heating values (HHV) of the untreated and torrefied fuels were determined using a Parr 6200 Bomb Calorimeter using the method laid out in the European Standard [25]. Using the HHV data with the mass yield method above, the energy yields for each torrefaction condition were calculated using the following equation:

\[ \eta_k = \eta_m \left( \frac{HHV_{\text{treated, dry}}}{HHV_{\text{untreated, dry}}} \right) \times 100 \]  

(2)

where \( HHV_{\text{treated, dry}} \) and \( HHV_{\text{untreated, dry}} \) correspond to the higher heating values of the torrefied and untreated fuels (dry basis) respectively.

2.2. Energy requirements for drying and torrefaction

2.2.1. Mass balance

To calculate the GHG emissions associated with the drying and torrefaction process, a mass and energy balance reflecting a real-life system was modelled based on experimental results obtained from the torrefaction of pine under each of the conditions listed in Table 1. Firstly, the initial mass flow in to the dryer was calculated as the mass of wet biomass required to produce 1000 kg of torrefied product. Using the results from proximate and ultimate analysis for untreated pine, the weight percent of each of the species C, H, N, O and ash were interpolated at 35% moisture to determine the flow of each of these species (plus moisture) entering the dryer. This was done firstly, to reflect the true composition of harvested biomass entering a dryer in a real-life system and secondly, as 35% was considered the minimum moisture content that could be achieved after storage in outdoor piles through natural drying post-harvest [26]. After drying at 110 °C, it was assumed that the moisture content of the biomass was reduced to 10% (with moisture exiting the dryer system) with no other changes to the fuel occurring. At 10% moisture, the biomass then enters the torrefier where it is torrefied under the conditions listed above. Once torrefied, the mass flow of each of the species (C, H, N, O, ash and moisture) exiting the torrefier were based on the ultimate and proximate analysis and dry mass yields obtained for the torrefied materials. To close the mass balance the volatile species and gases were calculated by difference.

The elemental composition of the volatiles stream was determined from the mass balance, however the individual species were estimated using the FC-Biomass model [27] to determine the latent heat of vaporisation for each species as discussed below. For this, heating rates, residence times and final temperatures and a specific fuel file were used as inputs. This provided yields of solids, condensable and gaseous products and their composition.

2.2.2. Energy balance

An energy balance was performed to determine the energy flows in to the torrefaction system and the energy contained in the torrefied product and volatiles stream. The HHVs were determined from bomb calorimetry and thus the LHV were calculated (using the methodology laid out in European Standard [25]) for the untreated pine (at 35% moisture) and each of the torrefied materials. In order to mimic a real-life system, the LHV of the fuels were used as the HHVs account for the energy that would be recovered from the condensation of steam upon combustion which is not applicable in industrial scenarios. The energy flow into the system was calculated using the LHV of the untreated biomass (at 35% moisture) and the mass flow into the dryer. The energy exiting the system was calculated using the LHV of the torrefied product (at each condition) and the mass flow out of the torrefier. The remaining energy by rule of conservation was assumed to all be contained in the volatile stream.

2.2.3. Energy requirements

Knowledge of the energy requirements for the overall torrefaction process is crucial in determining the GHG emissions associated with the process. The total energy required was split in to two stages: drying and torrefaction and calculated as the sum of the energies required to heat the dry biomass and moisture contained in the biomass (both accounting for the sensible energy requirements) and the latent heats of vaporisation of the moisture and volatile species. The calculations used to determine the energy requirements for drying and torrefaction are shown in the supplementary information.

The energy contained in the volatile stream was assumed to be all available for combustion at an efficiency of 85% to provide some of the heat demand required for drying and torrefaction. For each condition, the energy available in this stream was less than the energy required for drying and torrefaction and so the additional energy required was provided by a utility fuel: either wood chips (WC) or natural gas (NG). It is also important to note that the torrefaction reaction overall becomes exothermic above 250 °C [28] although some endothermic reactions take place.

2.3. Greenhouse gas emissions assessment

Life Cycle Assessment (LCA) studies the potential environmental impacts throughout a product's or system's life from raw material acquisition through production, use and disposal [29,30]. The purpose is to provide a holistic view of the emissions and resource requirements of a product system. In this paper the primary focus is on greenhouse gas (GHG) emissions which have been calculated in accordance with the EU Renewable Energy Directive (RED) [1], as updated for solid biomass [31]. Impacts of activities involved in torrefied wood pellet production include biomass cultivation, harvesting and collection, transportation, size reduction and screening, drying, torrefaction, pelletisation, storage, distribution and use of torrefied pellets (TP)/wood pellets (WP) to the end-user. The comprehensive view provided by LCA allows GHG emissions to be assessed on a whole system basis by life cycle stage.

To model GHG emissions for the torrefaction system it is necessary to define the key parameters of the supply chain. This is done by delineating the key resource and energy inputs and emissions from each life cycle stage. The following sub-sections provide a description of the TP production and use i.e. the process from pine forest through to electricity production. A detailed summary of the main inventory data and assumptions used in the study is provided in Table 2. This includes key parameters for the 4 torrefaction conditions considered from the experimental work (see Section 2.1).

The pine wood is assumed to be cultivated in Amory, Mississippi (MS), South-East USA [32]. Results are presented as the base case with an existing operational pellet facility chosen for the case study to assess actual supply chains. Further details are provided in the supplementary information.

2.3.1. Pine feedstock supply – cultivation, harvesting, chipping, and transport

Existing pine forests are well established and managed for wood supply. The total land required was calculated as the amount
incurring losses of 2.5% [36,37]. Wood chips are then transported to the forest roadside using diesel wood-chippers and chipped at the density of 385 kg/m³ [38]. Diesel is consumed for transportation assuming a transport distance being two thirds of the radius (see supplementary information).

2.3.2. Drying, torrefaction & storage

Once received at the torrefaction pellet facility, wood chip is stored on site before being dried to reduce the moisture content to 10%. The energy required for the drying and torrefaction processes are modelled using experimental data (see Section 2.1). Energy contained in the volatile species evolved during torrefaction (also known as ‘torgas’) are assumed to undergo combustion at an efficiency of 95% to provide some of the process energy required with the remaining energy required provided by a utility fuel. Two types of utility fuel are assessed in the results: WC and NG. Some electricity is also required for cooling, control equipment, and to meet the parasitic load.

2.3.3. Torrefied pellet production

After drying and torrefaction, the torrefied wood (TW) is cooled during subsequent processing operations [17]. This can occur at the elevated temperatures of 250 °C at which TW leaves the reactor [15]. Once cooled the TW enters a hammermill to reduce particle size to allow for pelleting using a pellet mill [40]. Both the hammermill and industrial pellet mill processes are assumed to be driven using a USA grid electricity mix [41]. Electricity demand for the of wood required to produce 100,000 tonnes of pellets and normalised to against the CV of the least torrefied fuel. This was done to account for changes in mass loss upon torrefaction with increase in CV. It is assumed that no fertilisers or pesticides are required, so the main emission source is the diesel consumed in cultivation and harvesting [33]. In accordance with the RED methodology, GHG emissions as a result of land use change (LUC) are not required to be calculated where a LUC has not occurred from 2008 [1,7]. Consequently, soil carbon and land-use change emissions are not considered in the inventory, but are discussed in Section 3.6.1. It is recognised that these are important issues for the carbon balance (also known as ‘torgas’) are assumed to undergo combustion at an efficiency of 95% to provide some of the process energy required with the remaining energy required provided by a utility fuel. Two types of utility fuel are assessed in the results: WC and NG. Some electricity is also required for cooling, control equipment, and to meet the parasitic load.

2.3.3. Torrefied pellet production

After drying and torrefaction, the torrefied wood (TW) is cooled to prevent combustion between the TW and atmospheric oxygen during subsequent processing operations [17]. This can occur at the elevated temperatures of 250 °C at which TW leaves the reactor [15]. Once cooled the TW enters a hammermill to reduce particle size to allow for pelleting using a pellet mill [40]. Both the hammermill and industrial pellet mill processes are assumed to be driven using a USA grid electricity mix [41]. Electricity demand for the
base case is assumed to be 15 kW h/t for the hammermill and 80 kW h/t for pelleting [13]. There are conflicting data in the literature regarding the energy requirements of pelleting torrefied biomass; hence this parameter is further assessed in the sensitivity analysis (see Section 3.5.2).

2.3.4. Transport & logistics to end-user

Once the TPs are produced they are exported to the UK using existing transport logistics and infrastructure. Pellets are assumed to be transported by road from Amory, MS to the port in Mobile, Alabama (AL) over a distance of 415 km. At the port, pellets are loaded onto an ocean-going vessel with 50 kt capacity. A product tanker transports the pellets for 8912 km to the Port of Hull, UK. From the port, pellets are transported to a Power Plant by road over a distance of 51 km. Further details of the transport logistics are provided in the supplementary information.

2.3.5. Electricity production

Pellets are assumed to be used for electricity production in a large scale power plant with 40% electrical efficiency [42].

2.3.6. Conventional wood pellet production

For comparative assessment, a conventional wood pellet (WP) supply chain was modelled using the same biomass as TP without the torrefaction stage. As shown in Table 2, most of the assumptions for WP remain the same as TP to allow for comparability. Key differences are summarised as:

- No torrefaction process involved, therefore all utility fuel from external sources.
- Additional energy requirement for grinding biomass prior to pelletisation.
- Lower calorific value and bulk density.

Key assumptions and input data for the GHG model described in Sections 2.3.1–2.3.6 are summarised in Table 2 by life cycle stage.

2.3.7. Scenarios and sensitivity analysis

Some aspects of this study were not based on experimental work or have uncertainties associated with them. To assess these further some different scenarios are considered in the results along with a sensitivity analysis of uncertain parameters. These can be summarised as follows:

- Use of torfags – results for the base case are assessed when no torgas is utilised (see Section 3.5.1).
- Electricity required for pelleting torrefied wood – low and high values from literature (see Section 3.5.2).
- Transport type (to port) – different options for land transport are considered (see Section 3.5.3).

3. Results

3.1. Torrefaction of North American pine

The mass yields (dry basis) for the torrefaction of pine under each condition are shown in Table 3. It can be seen that with increasing torrefaction severity, there is a decrease in the dry solid mass yield ranging from 90.7% to 72.2%. This decreasing yield is mainly attributed to loss of moisture and volatile material from the parent fuel through devolatilisation of the hemicellulose fractions which can become extensive as temperatures reach around 270 °C [14]. Under the temperatures and residence times of the more severe torrefaction conditions, 270-60 and 290-30, devolatilisation of the cellulose and lignin fractions can occur too resulting in even greater mass loss [14,48]. It can be seen that temperature has a greater effect on the mass yield than residence time when comparing these two parameters directly i.e. comparing 270-30 with 270-60 and 290-30, which is in agreement with other researchers [49,50]. The loss of reaction water and CO2 during torrefaction results in a loss of carbon, hydrogen and oxygen, while the light volatiles produced contain only small amounts carbon and relatively high amounts of hydrogen and oxygen resulting in a dark carbon enriched solid product whose lignocellulosic components are composed mainly of cellulose and lignin [51]. The results of the proximate and ultimate analysis shown in Table 3 highlight this effect in more detail. With torrefaction, the oxygen weight percent has decreased while the carbon content increases; with greater disparity between the untreated and torrefied fuels shown as the temperature and residence time increase. This effect is further shown from the results of proximate analysis where there is decrease in moisture content after torrefaction. The loss of moisture during torrefaction occurs via two main reactions: during the physical drying of wood (this performed prior to torrefaction) and via dehydration reactions of the organic components. It is worth noting that the moisture content of torrefied materials is not related to the torrefaction temperature and is due to intrinsic uptake of moisture after torrefaction. The results of the proximate analysis also show a decrease in the measured volatiles content and increase in fixed carbon content with increasing torrefaction, the latter an indication of lignin content [52]–highlighting an increase in this lignocellulosic component’s weight percent. There is also an increase in the ash weight percent – this concentration due to the loss of organic matter and decrease in the moisture content.

The loss of oxygen and enrichment of carbon upon torrefaction has a marked effect on the heating value of the fuels, as seen in Table 3. For the mildest treatment, 250-30, there is a small increase in HHV from 20.21 MJ/kg to 20.36 MJ/kg, which then increases to 23.49 MJ/kg for pine torrefied at 290-30. The HHV of fuels has great impact when determining its suitability for power generation as a higher HHV results in less biomass needed for a given amount of energy, however the HHV of a fuel cannot be solely isolated as an indicator for torrefaction optimisation. The energy yield is an important parameter that must be factored in and taken into account the energy lost as function of the HHV and the overall mass loss. Table 3 shows the energy yields for each condition, which range from 91.3% to 83.8% as torrefaction temperature and residence time increase. The energy yields are greater than the mass yields under each condition resulting in an overall increasing trend of energy densification for the torrefied fuels with increased HHVs. The increasing mass loss corresponds to a greater energy

| Table 3 | Results from the ultimate and proximate analysis for untreated and torrefied North American pine chips. |
|---------|--------------------------------------------------------------------------------------------------------|
|          | Untreated | 250-30 | 270-30 | 270-60 | 290-30 |
| C (%)a  | 49.68     | 51.88  | 52.57  | 54.12  | 54.95  |
| H (%)a  | 5.67      | 6.10   | 5.82   | 5.84   | 5.58   |
| N (%)a  | 0.13      | 0.12   | 0.11   | 0.10   | 0.09   |
| S (%)a  | 0.06      | N.D    | N.D    | N.D    | N.D    |
| O (%)b  | 44.46     | 41.89  | 41.49  | 39.54  | 39.37  |
| Moisture (%) | 7.08  | 2.43   | 1.86   | 1.13   | 2.02   |
| Volatiles (%)b | 83.78 | 81.66  | 79.64  | 76.35  | 72.78  |
| Ash (%)c | 0.34      | 0.45   | 0.35   | 0.47   | 0.55   |
| Fixed carbon (%)d | 15.89 | 17.89  | 20.01  | 23.18  | 26.66  |
| HHV (MJ kg⁻¹) | 20.21 | 20.36  | 21.22  | 22.09  | 23.49  |
| Mass yieldd | –      | 90.7  | 85.0   | 76.7   | 72.2   |
| Energy yieldd | –    | 91.3  | 89.2   | 83.8   | 84.0   |

a Dry-ash free.
b Dry-basis.
c As-received.
d Calculated by difference.
loss despite the increase in HHV, which becomes significant when scaling up and economics of processes are considered. Typical mass and energy yields for torrefaction are often cited as 70% and 90% respectively [14] and if these criteria were to be considered as optimal, the two mildest conditions: 250-30 and 270-30 are within acceptable limits. The most severe conditions: 270-60 and 290-30 however may be considered inefficient as while the mass yields are within range, just over 15% of the original energy content of the fuel is lost resulting in lower energy yields despite the marked improvement in HHV. The energy lost during torrefaction has further implications when analysing the potential for utilising energy in the volatile stream (‘torgas’) for heat to power the torrefaction process, which will be discussed later in the text.

3.2. Composition of volatile species determined using FG-Biomass

The composition and yields of the species contained in the volatiles stream were modelled using the FG-Biomass model and are shown as Fig. 1. It can be seen that for each species, the yields increase with increasing torrefaction severity. Reaction water represents the highest yield for all conditions followed by carbon dioxide then acetic acid. These results are comparable with the volatile species quantified experimentally by Prins et al. [53] in which the authors ascribe the formation of these species to occur as a result of decomposition of the hemicellulose fraction. The small amounts of carbon monoxide present, as noted by Prins et al., cannot be explained by decomposition reactions involving the cell wall species. The authors thus attribute the formation of carbon monoxide to occur as a result of the reaction of carbon dioxide and steam with char as temperatures increase [53].

3.3. Land required

The results for the amount of land required for pellet production and additional wood for utility fuel are shown in Table 4.

The land required for production of torrefied pellets increases with the trend 250-30 < 270-30 < 290-30 < 270-60. As torrefaction severity increases there is greater mass loss which would correspond to more input fuel required and thus more land to produce the same amount of feedstock. However, as torrefaction increases the CV of the resultant fuel, there is a compensation effect as there is more energy contained in the most torrefied fuels and accounting for this increase with respect to mass loss allows the land requirements to be calculated assuming the same energy output in the torrefied pellets. This is evident when comparing the land required for pellet production for conditions 270-60 and 290-30 as there is greater input mass required to make 1 ton of torrefied pellets for condition 290-30 however less land required as a result of higher CV (Table 3). The additional mass required to account for using this feedstock as a utility fuel is also shown in Table 4. Condition 250-30 requires more additional utility fuel as there is less energy available in the volatile stream to be used for combustion to heat the torrefaction process. In combining the land requirements for pellet production and utility fuel, condition 270-30 requires the least amount of land overall. When comparing the land required for untreated wood pellets however, it can be seen that less land is required when compared with each torrefaction condition.

3.4. Greenhouse gas emissions assessment

Greenhouse gas (GHG) emission results are presented for the base case using the assumptions outlined in Section 2.3. For each of the four torrefaction conditions two options were considered for utility fuel being wood chips (WC) and natural gas (NG) to produce torrefied pellets. Results are also presented for a conventional wood pellet (WP) for comparison purposes. Fig. 2 shows the results for these different pellets broken down into the 9 life cycle stages.

Fig. 2 shows that treatment 250-30 results in 29.4 CO₂e/MJ which has the highest emissions when compared to other TPs on a ‘per MJ’ basis. The primary reason for this is the limited amount of torgas available from the volatiles from the less severe conditions which result in a fuel with lower calorific value (CV). By increasing the temperature by 20 °C the emissions for 270-30 are reduced to 27.9 gCO₂e/MJ for WC and 38.8 gCO₂e/MJ for NG. Even greater GHG savings are obtained when the torrefaction severity increases to conditions 270-60 and 290-30. This trend is observed due to an increased CV of the torgas produced as a result of longer residence times and temperature respectively, requiring less additional utility fuel. While the reduction in consumption of additional fuel is desirable from a GHG emissions perspective, the additional energy available in the torgas stream for the more severe conditions is available at the expense of the energy contained in the parent fuel as discussed above. The parameters with which torrefaction optimisation are to be ascribed must therefore be clearly defined when making assessments of GHG emissions. In this instance, if the mass and energy yields are to remain within traditional guidelines, torrefaction under conditions 270-60 and.

Table 4

| Mass input required to make 1000 kg of pellets (kg) | 250-30 | 270-30 | 270-60 | 290-30 | WP |
|-----------------------------------------------|--------|--------|--------|--------|----|
| Land required for 1656 | 1777 | 1983 | 2086 | 1385 |
| Land required for wood pellets* (ha) | 26,805 | 27,501 | 28,930 | 28,726 | 24,730 |
| Land required for WC utility fuel (ha) | 2639 | 1811 | 577 | 776 | 1974 |
| Total land required (ha) | 29,444 | 29,313 | 29,507 | 29,503 | 26,705 |

* Normalised for CV.
290-30 may be considered uneconomic as greater amounts of feedstock will be required, which may result in potential rejection despite lower overall emissions. The emissions associated with the utility fuel subsequently play a crucial role when looking at GHG emissions – if a higher proportion of heat for torrefaction is sourced externally. Differences in results are less pronounced when WC are used for drying and torrefaction, showing that the torrefaction condition is more significant (from a GHG emissions perspective) when NG is used as utility fuel. Combustion of NG has much higher emissions factor than WC as a fossil derived energy carrier, whereas carbon emitted from WC is considered to be biogenic with an emissions factor close to zero [41].

With biomass sustainability criteria becoming increasingly important, the GHG emissions from biomass electricity are required to meet thresholds of 79.2 gCO₂e/MJ of electricity delivered, a saving of 60% against the EU fossil average for electricity generation [54]. This threshold has been implemented following recommendations from the EU [31], and under current UK legislation will reduce over time [54]. All pellets presented in Fig. 3 would meet the existing GHG criteria, and show that using biomass rather than fossil fuels for utility fuel is crucial to maximise GHG savings of the supply chain.

All 4 TPs produce lower GHG emissions than conventional WP showing that despite the additional processing step, the use of torgas and increased CV of the TPs lowers their GHG emissions on a ‘per MJ’ basis. When the different life cycle stages of production are assessed, it is apparent that cultivation, harvesting, chipping, and transport to the pellet plant (collectively grouped as ‘feedstock supply’) have broadly the same emissions for all pellets considered. Feedstock supply contributes approximately 2.9–3.8 gCO₂e/MJ of electricity delivered to each pellet supply chain and is therefore not further analysed here. It should however be reiterated that emissions from biomass feedstock supply can vary substantially depending on wider factors such as land use change, carbon debt, soil carbon and system boundary definition, and also specific variables; for example fertiliser inputs, fuel use, processing, and transportation distance [33,55–60].

The contribution of drying and torrefaction are of crucial importance when considering the life cycle GHG emissions, particularly with regard to the choice of utility fuel. Fig. 3 portrays that emissions from drying and torrefaction could be as low as 0.5 gCO₂e/MJ of electricity delivered (<2% of total) for 290-30 (WC) or as high as 14.9 gCO₂e/MJ of electricity delivered (~34% of total) for 250-30 (NG). For WPs the contribution from drying using WC and NG is 2.9% and 27.7% of the total respectively.

The emissions from pellet production derive from the energy required to grind the wood to smaller particles before pelleting followed by compression and extrusion in the pellet press. During torrefaction, decomposition of the lignocellulosic components in biomass occurs, with hemicellulose the most reactive under
thermal treatment. As this polysugar provides structural integrity in the pine wood cells, its full or partial degradation leads to improved grindability and thus lower amounts of energy required in particle size reduction. The emissions for pellet production for the TPs range from 5.7 to 6.7 gCO₂e/MJ while the emissions for production of WP (with no torrefaction) are 10.8 gCO₂e/MJ. The assumptions for electricity use vary in the literature and are therefore assessed further in the sensitivity analysis (Section 3.5.2).

Emissions from road transportation of pellets to the shipping port reduce with higher torrefaction severity due to the increased cv and bulk density. The contribution of road transport to the port for WP is the highest at 4.4 gCO₂e/MJ with this reducing to the lowest of 3.4 gCO₂e/MJ for 290-30. Utility fuel type influences emissions up to the point of pellet production; however for the transportation logistics it is primarily the energy content of the fuel that determines the GHG balance. For densified biomass such as pellets, transport is usually mass restricted whereas unprocessed biomass (e.g. wood chips) with higher moisture and lower bulk density, the volume is frequently the limiting factor.

Shipping is the biggest emission source for all scenarios, except for 250-30 where drying and torrefaction is larger when NG is used. Shipping emissions reduce as the calorific value of the pellet increases therefore 290-30 has lowest emissions from transport with WP the highest, representing one of the key potential advantages of torrefaction. Emissions for drying are higher for WP due to the assumption that no torgas is available to reduce demand for utility fuel.

3.5. Sensitivity analysis

For the sensitivity analysis 3 main areas were highlighted for additional assessment. Feedstock supply was considered outside the scope for further analysis as all pellets assessed have the same emissions up to the point of delivery to the pellet processing plant. The sensitivity cases therefore focus on (i) the use of torgas; (ii) electricity required for pelleting torrefied wood; (iii) transport type (to port).

3.5.1. Use of torgas

Making use of the torgas is of key importance when assessing the GHG emissions from different torrefaction conditions [18]. There are limits to the degree of mass and energy loss that should occur in torrefaction that will affect the amount of energy contained in the volatile stream as discussed in Sections 3.1 and 3.4. Higher temperature and longer residence time means that more energy is available in the torgas reducing the utility fuel requirement. Nonetheless, the more severe torrefaction conditions presented here result in greater mass loss and consequently more biomass is required, thereby increasing the land required and associated economic cost. While the energy available in the torgas is modelled in this study and assumed to be all available for combustion (at 95% efficiency) its application in real-life scenarios is accompanied with several design and process considerations which would be factored in the event of a pilot or production-scale torrefaction plant being built. Such considerations include whether the combustion of torgas provides heat directly or indirectly to the incoming fuel where in each case the fuel to be torrefied either comes in contact with the heat carrier or is heated via a physical separation (e.g. a wall) respectively [14]. Other design considerations could also include utilising the heat from the torrefied product exiting the torrefier to reduce the amount of additional utility fuel required. The design and considerations mentioned here are beyond the scope of this study, however the impact of using no torgas was considered as part of the sensitivity analysis and to demonstrate its significance, emissions were calculated for a scenario with no torgas available. Fig. 3 depicts how the calculated GHG emissions change for ‘drying and torrefaction’ when all of the thermal energy requirements are assumed to be supplied by utility fuel (either WC or NG) with zero torgas. It is observed that not using torgas makes results for NG drying much higher than the base case results, particularly for 270-60 and 290-30. All torrefaction cases show similar results when no torgas is available, with results comparable to WP when NG is used. Using wood chips for ‘drying and torrefaction’ has less more of an impact on results with these out-performing WPs under each condition.

Table 5 summarises how the results change for the 2 scenarios, with and without torgas, for the different pellets.

3.5.2. Electricity required for pelleting torrefied wood

As mentioned in Section 2.3.3, there is a lack of agreement in the literature regarding data on the electricity required for pellet production, which will vary depending on the nature of the feedstock, degree of torrefaction, and type of mill and pellet pressed used to determine consumption. It is generally known that less energy is required to reduce torrefied wood chips to smaller particles prior to pelleting than untreated wood chips since torrefaction can improve the grindability behaviour of fuels and so has an impact on the overall electricity consumption of the process when torrefaction is combined with pelleting [48]. However, uncertainties lie in the energy required for compression and extrusion of pellets from torrefied biomass. Some researchers, such as Stelte et al. [61], argue that the loss of moisture (which acts as a plasticizer) and extractives during torrefaction increase friction in the channel

| Table 5 | Results for the sensitivity analysis for no torgas. |
|---------|---------------------------------------------------|
|         | Total GHG emissions gCO₂e/MJ(electricity)          | Emissions from drying and torrefaction gCO₂e/MJ(electricity) |
|         | With torgas | No torgas | Difference (%) | With torgas | % of total (%) | No torgas | % of total (%) |
|         |             |           |               |             |               |           |               |
| Wood chips |             |           |               |             |               |           |               |
| 250-30   | 29.4        | 29.6      | 0.7           | 1.1         | 3.8           | 1.3       | 4.4           |
| 270-30   | 27.9        | 28.4      | 1.7           | 0.9         | 3.3           | 1.4       | 4.9           |
| 290-30   | 25.9        | 26.9      | 3.9           | 0.4         | 1.7           | 1.4       | 5.3           |
| 290-30   | 24.5        | 25.4      | 3.7           | 0.5         | 2.0           | 1.4       | 5.5           |
| Wood Pellet | N.A        | 35.6      | N.A           | N.A         | N.A           | 1.1       | 3.0           |
| Natural gas |             |           |               |             |               |           |               |
| 250-30   | 43.1        | 46.6      | 8.1           | 17.6        | 38.9          | 18.0      | 39.4          |
| 270-30   | 38.8        | 46.7      | 20.3          | 11.6        | 30.5          | 19.7      | 42.2          |
| 290-30   | 28.9        | 44.3      | 53.3          | 3.7         | 12.7          | 20.7      | 44.8          |
| Wood Pellet | N.A        | 47.8      | N.A           | N.A         | N.A           | 13.2      | 27.7          |

| N.A = not applicable. |
press resulting in higher pelletizing pressures and subsequent energy uptake of the mill (which increases as torrefaction severity increases). In agreement with this notion is the study by Li et al. [62] who attributed reduced plasticity, and therefore increased extrusion and compression in the pellet press, to the degradation of hemicellulose and lignin during torrefaction thus resulting in greater energy requirements when compared to pelletizing untreated wood. Similar trends have been reported from pilot-scale pelletizing of spruce torrefied at 270 °C and 300 °C for 16.5 min by Larsson et al. [63], where 100% more energy was required for pelletling torrefied material when compared to pelletting of untreated fuel. Moreover, it was also found that torrefied pellets were less durable and had only comparable bulk densities to untreated pellets.

In contrast, Bergman et al. [64] report lower overall power consumptions are required for pelletling torrefied biomass when compared to untreated biomass. The authors also report higher bulk densities for torrefied pellets compared with conventional pellets (750–850 kg/m³ and 500–650 kg/m³ respectively) and that the torrefied pellets obtained showed improved mechanical strength with crushing tests demonstrating that torrefied pellets could withstand 1.5–2 times more force than conventional wood pellets. The authors attribute this to alterations of fatty structures during torrefaction, which serve as binding agents, as well increased lignin weight percent providing mechanical strength. The role of lignin in WPs is very important as it acts as a binder in pellet production and contributes to pellet mechanical strength. It is generally agreed that upon heating through compaction, the lignin in wood particles, with aid of moisture, undergoes softening and transitions from a ‘glassy to rubbery’ composition acting as a glue between particles via hydrogen bonding on the surface with hemicellulose [13,19]. Although there are different views on the role of lignin in torrefied pellet production, it is known that in the case of severely torrefied materials (i.e. T > 280 °C) the resultant pellets have less mechanical strength than those torrefied under milder conditions (e.g. [61]) in some cases not producing viable pellets at all. It has been suggested that only low molecular weight polymers are involved in glass transition and binding in wood pellets and as these can degrade during torrefaction, pellets produced under certain torrefaction conditions lack mechanical strength and durability [62]. These problems may be overcome from increasing the die temperature in the pellet press to encourage the glass transition of higher weight lignin polymers or the addition of a binding agent; however these will ultimately have implications on energy consumption and subsequent GHG emissions.

Furthermore, the values for electricity consumption found in the literature are based on laboratory-scale mills and single pellet presses, which are not synonymous with large-scale industrials mills. As pointed out by Jarvinen and Agar [13], industrial data are scarce as pelletling of torrefied wood on large scale is often performed internally; requiring large amounts of feedstock that are not often produced in academic institutions. This leads to a gap in the information available resulting in the use of laboratory or semi-industrial scale data, which may not reflect real-life scenarios and affect the results of GHG emissions assessments.

Due to the lack of large scale data and issues discussed above, a sensitivity analysis for the electricity consumption for pelletling torrefied wood was deemed necessary as any uncertainties may have a sizeable impact on associated GHG emissions. The results of the sensitivity analysis for the electricity required to pelletise torrefied wood are shown in Fig. 4. The low and high case scenarios were taken from Batidzirai et al. [65] and are 18 kW h/t and 395 kW h/t respectively. The results show little change in overall GHG emissions for each torrefaction condition under the low case scenarios (18 kW h/t) when WCs are used with values ranging between 19.9 and 23.5 gCO₂e/MJₑlectrificey. When NG is utilised the emissions range from 24.3 gCO₂e/MJₑlectrificey to 37.0 gCO₂e/MJₑlectrificey for biomass torrefied at 270-60 and 250-30, respectively. Under the high case scenario (395 kW h/t) a similar trend is observed. However, the emissions using WC range from 42.4 to 49.5 gCO₂e/MJₑlectrificey while the NG emissions are much higher ranging from 46.8 to 66.0 gCO₂e/MJₑlectrificey. Comparing to conventional WP emissions, when WCs are used, the low case scenario outperforms the WP emissions—although under the high scenario the reverse is shown. When comparing TP and WP supply chain emissions where NG is used as utility fuel, the low case scenarios for all TPs outperform WP emissions. Under the high case scenario, only the 270-60 and 290-30 life-cycle emissions outperform the WP emissions.

3.5.3. Transport type (to port)

The base case assumes that road transport is used for transporting pellets to/from transatlantic shipping ports; however in several locations alternative options include freight-trains or inland water barges. In particular, the pellet facility chosen for this case study uses inland barges [32]. The sensitivity analysis results for wood chip (WC) only, assuming a distance of 415 km is displayed in Table 6. Fuel use and emission factors are taken from Biograce [39]. These results show that reductions in GHG emissions of 7.0–8.6% are achievable with rail (electric), rail (diesel), and inland water barges. Over the distance of 415 km, the GHG emissions from transport using alternative transport to road trucks can reduce from 3.4–4.4 gCO₂e/MJₑlectrificey (12.4–13.9% of the total) to 1.4–1.9 gCO₂e/MJₑlectrificey (4.9–6.4% of the total).

3.6. Other aspects

3.6.1. Land use change and soil carbon

The life-cycle emissions determined in this study adopt the RED methodology which considers the emissions associated with harvesting, processing, transport and combustion and consequently do not consider the emissions associated with land use change (LUC)/indirect land use change (ILUC) prior to 2008 [66]. The inclusion of LUC and ILUC within the system boundaries is often challenging as specific data pertaining to LUC/ILUC is difficult to determine with certainty [34]. It can also be difficult to relate changes in LUC/ILUC with bioenergy systems being assessed i.e. the model outputs measuring carbon stocks are not strictly related to the functional units used for bioenergy systems; in this instance gCO₂e/MJₑlectrificey [55].

Several studies have attempted to include changes in carbon stock within the system boundaries of bioenergy LCA e.g.
3.6.2. Emissions from outdoor drying

Emissions can also arise from storage of biomass that can contribute to GHG emissions including CO$_2$, nitrous oxide (N$_2$O) and methane (CH$_4$). The gaseous emissions from storage are linked to dry matter loss which occurs as a result of degradation of the wood. The extent of degradation depends on the nature of the feedstock, storage environment and moisture content [70,71]. CO$_2$ emissions from wood can occur from thermal oxidation and aerobic and anaerobic biodegradation, while action of microorganisms in anaerobic conditions results in CH$_4$ evolution [71]. N$_2$O emissions occur as the end product of incomplete ammonium oxidation of incomplete denitrification [70]. He et al. report on the emissions of CO$_2$ and CH$_4$ from Canadian Douglas fir branches with higher emissions for both gases at higher temperatures (35 °C when compared to 15 °C) and peak concentrations of 138,000 ppm and 1500 ppm respectively, most likely as a result of increased microbial activity at higher temperatures [71]. The authors also noted a decrease in oxygen concentrations to 1–2% after 10 days storage. Theoretical methane and nitrous oxides losses from wood residues were calculated by Wihersaari who calculated daily emissions rates of 24 g/m$^3$ and 0.6 g/m$^3$ for methane and nitrous oxide respectively [71]. The conclusion of this study was that forest residue should be utilised as quickly as possible to avoid emissions from this source. These emissions may present an issue when natural of drying wood occurs, particularly in the summer months where the outdoor climate is warmer leading to increased microbial activity.

4. Concluding remarks

The results of the torrefaction of North American pine and an assessment of life-cycle GHG emissions for torrefied pellet production are presented. It can be seen that torrefaction improves the fuel properties of pine: the fixed carbon and HHV increases while the moisture and volatiles contents decrease. These changes become more pronounced as torrefaction severity increases. Results of the GHG emissions assessment shows potential GHG savings from conventional wood pellets based on the experimental results and assumptions described in this paper. The largest emissions by life cycle stage are caused by shipping followed by the torrefaction and drying stages while several stages of production, i.e. cultivation, transport to pellet facility, have a relatively small contribution to overall supply chains emissions. The use of torgas is critical for emissions savings where natural gas is used for utility fuel owing to the greater life-cycle GHG emissions and thus using...

### Table 6
Results for the transport sensitivity analysis.

| Transport mode | Truck | Rail | Inland water barge |
|----------------|-------|------|--------------------|
| Fuel utilised  | Diesel| Electricity| Heavy fuel oil |
| 250-30 Transport emissions (gCO$_2$/MJ$_{electricity}$) | 3.9 | 1.5 | 1.7 |
| Total supply chain emissions (gCO$_2$/MJ$_{electricity}$) | 28.8 | 26.4 | 26.6 |
| Contribution of transport to supply chain | 13.5% | 5.7% | 6.4% |
| Change from base case | – | –8.4% | –7.7% |
| 270-30 Transport emissions (gCO$_2$/MJ$_{electricity}$) | 3.8 | 1.4 | 1.6 |
| Total supply chain emissions (gCO$_2$/MJ$_{electricity}$) | 27.9 | 25.5 | 25.7 |
| Contribution of transport to supply chain | 13.6% | 5.5% | 6.2% |
| Change from base case | – | –8.6% | –7.9% |
| 270-60 Transport emissions (gCO$_2$/MJ$_{electricity}$) | 3.6 | 1.4 | 1.5 |
| Total supply chain emissions (gCO$_2$/MJ$_{electricity}$) | 25.9 | 23.7 | 23.8 |
| Contribution of transport to supply chain | 13.9% | 5.9% | 6.3% |
| Change from base case | – | –8.5% | –8.1% |
| 290-30 Transport emissions (gCO$_2$/MJ$_{electricity}$) | 3.4 | 1.3 | 1.4 |
| Total supply chain emissions (gCO$_2$/MJ$_{electricity}$) | 24.5 | 22.4 | 22.5 |
| Contribution of transport to supply chain | 13.9% | 5.8% | 6.2% |
| Change from base case | – | –8.5% | –8.1% |
| Wood pellet Transport emissions (gCO$_2$/MJ$_{electricity}$) | 4.4 | 1.6 | 1.9 |
| Total supply chain emissions (gCO$_2$/MJ$_{electricity}$) | 35.6 | 32.8 | 33.1 |
| Contribution of transport to supply chain | 12.4% | 4.9% | 5.7% |
| Change from base case | – | –7.8% | –7.0% |
wood chips as utility fuel is preferred to keep life-cycle emissions as low as possible.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.enconman.2016.01.006.

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