



Techno-economic and environmental assessment of decentralized pyrolysis for crop residue management: Rice and wheat cultivation system in India

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ARTICLE INFO

Handling Editor: Zhifu Mi

Keywords:

Crop straw
Bio-oil
Biochar application
Global warming potential
Biorefinery
Biomass pretreatment

ABSTRACT

The current study evaluates a decentralized biorefinery's economic and environmental performance, which uses two-step pyrolysis for converting rice and wheat straw to bio-oil and biochar. The biorefinery was located in Punjab (India), where open burning of residue is prevalent. The decentralized biorefinery was evaluated regarding the energy required for pyrolysis, product yield and applications, and global warming potential (GWP). Pyrolysis of unwashed, water-washed, and acid-washed straws was performed. The net GWP for pyrolysis of unwashed rice straw was $-121 \text{ kg CO}_{2\text{eq}}/\text{ton}$ when biochar was used as a carbon sink. But pyrolysis of water-washed ($159 \text{ kg CO}_{2\text{eq}}/\text{ton}$) and acid-washed rice straw ($311 \text{ kg CO}_{2\text{eq}}/\text{ton}$) was a net contributor to GWP, which was undesirable. Similar trends were observed for wheat straw pyrolysis.

The GWP of washed rice and wheat straw pyrolysis was higher than unwashed straw because 37–50% biochar generated was used for drying the washed straw, leaving less biochar for further application. Amongst the biochar applications considered, its use as a carbon sink offered more GWP reduction than its use as a substitute for coal in power and heat generation. The net GWP for direct transfer of residues to a centralized refinery for pyrolysis was $-75 \text{ kg CO}_{2\text{eq}}/\text{ton}$ for rice straw and $-384 \text{ kg CO}_{2\text{eq}}/\text{ton}$ for wheat straw. Therefore, it was environmentally beneficial to treat biomass locally rather than in a centralized unit. Finally, the minimum selling price for biochar was calculated to be 172–623 USD/ton, which was within the range of commercial biochar price. Therefore, the proposed biorefinery was expected to be environmentally and economically viable with an appropriate selection of pretreatment options and end-uses of the products.

1. Introduction

Crop residues are an abundantly available lignocellulosic feedstock that can provide multiple value-added products in a biorefinery (Kamm, 2007). However, approximately 24% of the generated residue is burned in the fields in India (Ravindra et al., 2019). The open burning results in nearly 211 Tg (Teragram, 10^{12}g) of $\text{CO}_{2\text{eq}}$ greenhouse gases annually and 824 Gg (Gigagram, 10^9 g) of particulate matter (Ravindra et al., 2019). Further, open burning increases fertilizer and pesticide requirements to maintain productivity due to heat damage to soil microbial health (Cassou, 2018).

Governments' regulatory efforts have not significantly reduced residue burning (Kaushal, 2020; Shyamsundar et al., 2019). Rice straw and wheat straw burning are prevalent in India due to limited time for land

clearing between cropping cycles, prohibitive costs of land-clearing equipment, and low to no profit margins in the sale of residues (Cassou, 2018). Hence, this practice needs to be addressed by incentivizing farmers with economical and environmentally viable solutions through joint actions from the government and private sectors.

Globally, integrated biorefineries which combine decentralized pyrolysis and centralized refining of bio-oil have been proposed to address issues of open burning (Cherubini and Ulgiati, 2010). Such biorefineries can be operated on farms, saving time/energy for residue transportation, meeting the local energy demand, and increasing farmer incomes through value-addition to the residues.

Recent developments in pyrolysis-based residue treatment have been listed in the supporting information (Table S1). These studies show that a pyrolysis unit can be a self-sustaining that is the required process heat

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<https://doi.org/10.1016/j.jclepro.2022.132998>

Received 17 April 2022; Received in revised form 5 June 2022; Accepted 30 June 2022

Available online 8 July 2022

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can be supplied by burning part of the generated biochar or pyrolysis gases (Morgano et al., 2018). Pyrolysis also brings environmental savings because biochar can be used for carbon sequestration or a fossil coal substitute (Peters et al., 2015). Further, there are environmental savings from the avoided burning of residues (Chhabra et al., 2021) and profits from the production of higher-value products. For instance, bio-oil-derived levoglucosan showed a 75% lower global warming potential than its petrochemical alternative (Zheng et al., 2018).

Despite the benefits, there are conflicting views on the economic feasibility of decentralized biorefineries compared to fixed centralized refineries. Xin et al. (2021) showed that although the capital investment on a fixed 100-ton facility (US\$ 1.2 million) was higher than a mobile 25-ton unit (US\$ 0.5 million), the profits from a fixed unit exceeded the mobile unit due to a larger scale of production. In contrast, Chen et al. (2018) showed that the fixed plant (4000 kg/h) had a lower initial investment, but the mobile plant (100 kg/h) had a lower biofuel production cost and larger profit margins. The mobile system was profitable in the 6th year, while the fixed system took 7 years. Hence, performing regional and biomass-specific environmental and economic assessments is essential.

This study assessed the environmental performance of a decentralized two-step pyrolysis-based biorefinery with rice or wheat straw as feed, operating in Punjab (India), where crop residue burning is prevalent. Stepwise pyrolysis involves heating biomass in discrete steps to concentrate chemicals into separate fractions (Bhatnagar et al., 2020). The bio-oil is obtained according to the thermal degradation temperatures of biopolymers (hemicellulose, cellulose, and lignin) in the biomass. The pyrolysis conditions (temperature, heating rate, reactor design) and biomass determine the product quality. While, in the current study, biomass composition could not be controlled during the cultivation stage, it was altered using water-washing and acid-washing before pyrolysis. The biochar obtained in the pyrolysis process was used for soil amendment or as a substitute for coal. The bio-oil may be further separated into bulk and specialty chemicals like furfural, acetic acid, levoglucosan, and phenolic resin (Pinheiro Pires et al., 2019). However, the chemical recovery potential was not evaluated in this study. Another aim of the current study was to propose a selling price for the biochar generated from straw and compare its competitiveness with commercially available biochar sold in the Indian market, thereby assessing the economic feasibility of the proposed plant.

The current study would prove significant owing to the lack of detailed reports published on the extent to which decentralized pyrolysis

may address the issue of open burning, especially in the study area (Punjab, India). Further, the authors used experimental results and existing knowledge to model the biorefinery and identify factors that contribute to the environmental performance of a decentralized biorefinery specific to a region. The environmental performance of the proposed decentralized biorefinery was also compared with a fixed centralized biorefinery where straw was directly transported for pyrolysis, and with other techniques for straw use such as biological treatment for bioethanol or biogas synthesis and incineration for power. A sensitivity analysis was performed to determine which parameters affect most the complete techno-economic-environmental performance.

2. Methodology

2.1. The geographical boundary for the biorefinery

The biorefinery proposed in this study is in Punjab, India's major rice and wheat-producing state. The rice grain produced is 12.8 million tons/year with a yield of 4132 kg/ha, and wheat grain is 18.2 million tons/year with a yield of 5188 kg/ha (Directorate of Economics and Statistics, 2019). Due to the limited accessibility, it was assumed that only 50% of the residual straw generated was available for pyrolysis (Singh, 2017).

Based on the number of recorded fire incidents, 10 areas (districts) were selected in Punjab (Fig. 1), where pyrolysis units could be set up. The bio-oil produced would be transported to a chemical refinery in the Bathinda district, where an existing 11.3 million tons/year petroleum refinery could be retrofitted for bio-oil refining. Distances between the 10 districts and the chemical refinery were calculated based on the traffic map, and the median distance (85.3 km) to the refinery was used for the assessment. When comparing the direct transport of field straw to a centralized plant for pyrolysis and chemical separation, the same distances were used.

2.2. Configuration of pyrolysis unit based on regional specifications

The pyrolysis unit was estimated to operate 328 days a year and have a capacity for pyrolysis of 10 tons of dry straw per day. The annual straw requirement for one pyrolysis unit was calculated to be 3.28 kilotons (kt).

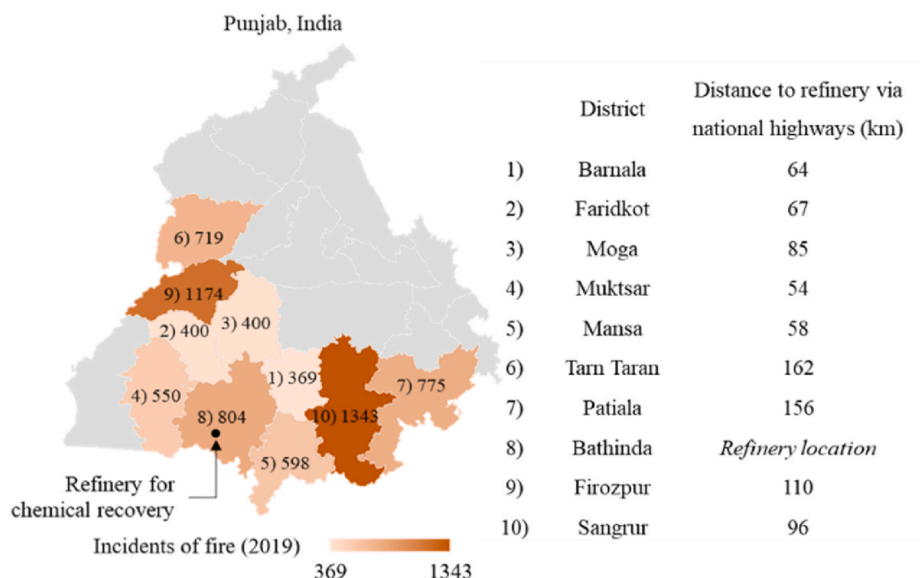


Fig. 1. Geographical distribution of decentralized units.

2.3. Life cycle assessment framework

Life cycle assessment (LCA) is a standardized technique often used to calculate the environmental impact of a product, process, or service. The LCA framework provided in ISO14040 and ISO14044 standards (ISO, 2006a; 2006b), was followed in this study.

2.3.1. Goal and scope

The current study reports the environmental impact of pyrolyzing rice straw (RS) and wheat straw (WS). Three cases of pyrolysis (Fig. 2) were evaluated. The first was pyrolysis of untreated straw, the second was pyrolysis of water-washed straw, and the third was pyrolysis of acid-washed straw.

A cradle-to-gate system boundary was used for crop residue treatment. The first step of the system boundary was the cultivation of straw (cradle), and the last step was the transport of bio-oil to the refinery (gate) for chemical recovery. The chemical recovery process from bio-oil was not included in the study. A system expansion approach was followed to include the credits from avoided products or processes. Wherever possible, the data used was specific to the study region. Government reports with national statistics provided the data for cultivation and farm operations. Information regarding machinery, fertilizers, chemicals, and fuel use was obtained from the EcoInvent 3.8 database. Previously published work (Bhatnagar et al., 2020, 2022; Cen et al., 2019) provided inventory data for pyrolysis, product yields, and energy and auxiliaries consumed. The emission inventory for each subsystem of the biorefinery was estimated from reported emission factors. The infrastructure-related emissions were not considered in the study. Emissions from the combustion of biochar and straw were considered biogenic. Impacts of land-use changes are not addressed. The LCA was performed using SimaPro 9.3.0.3. Material and energy requirements for all subsystems were evaluated using the reference flow or functional unit of 1-ton size-reduced dry straw (particle size: 1–3 cm) to be pyrolyzed.

2.3.2. Life cycle inventory for biorefinery case studies

The biorefinery was divided into seven subsystems: cultivation, baling, transport to the collection center, size reduction (optional washing and drying), pyrolysis, char application, and bio-oil transport to the chemical refinery.

2.3.2.1. Cultivation. The material and emissions inventory for the cultivation is provided in the supporting information (Table S2). Economic allocation was used to distribute the impacts of the cultivation of straw. The allocation was based on the minimum support price offered to farmers (Directorate of Economics and Statistics, 2019) for rice (INR 1732/100 kg) and wheat grain (INR 1855/100 kg). Duncan et al. (2020) estimated the minimum selling price of straw based on composition and seasonal variation as INR 2.46/kg for RS and INR 2.95/kg for WS. A crop to residue ratio (w/w) of 1:1.5 was assumed for rice and 1:1.75 for wheat crops (Sahu et al., 2021).

2.3.2.2. Baling. The operations in the field remained similar in all biorefinery case studies. The residual straw on the field was collected using balers. The data for baling and associated agricultural activities were obtained from field studies (ICAR, 2008), and the inventory for making 1 bale is provided in the supporting information (Table S3).

2.3.2.3. Transport to a collection center. After baling, the residues were transported from the field to collection centers and unloaded there. The pretreatment and pyrolysis activities were performed at the same collection center. It is assumed that residues are transported a distance of 10 km in tractors with 2-ton capacity trolleys. The mileage of a tractor is 4.5 km/dm³ of diesel (Soam et al., 2017), with estimated transportation losses of 0.5% by weight (Cardoen et al., 2015). Diesel requirements for loading and unloading operations are about 0.1 dm³/ton of biomass (Sreekumar et al., 2020). The diesel engines operate under the EURO IV emission regime (International Council on Clean Transportation, 2018). The inventory related to transport and

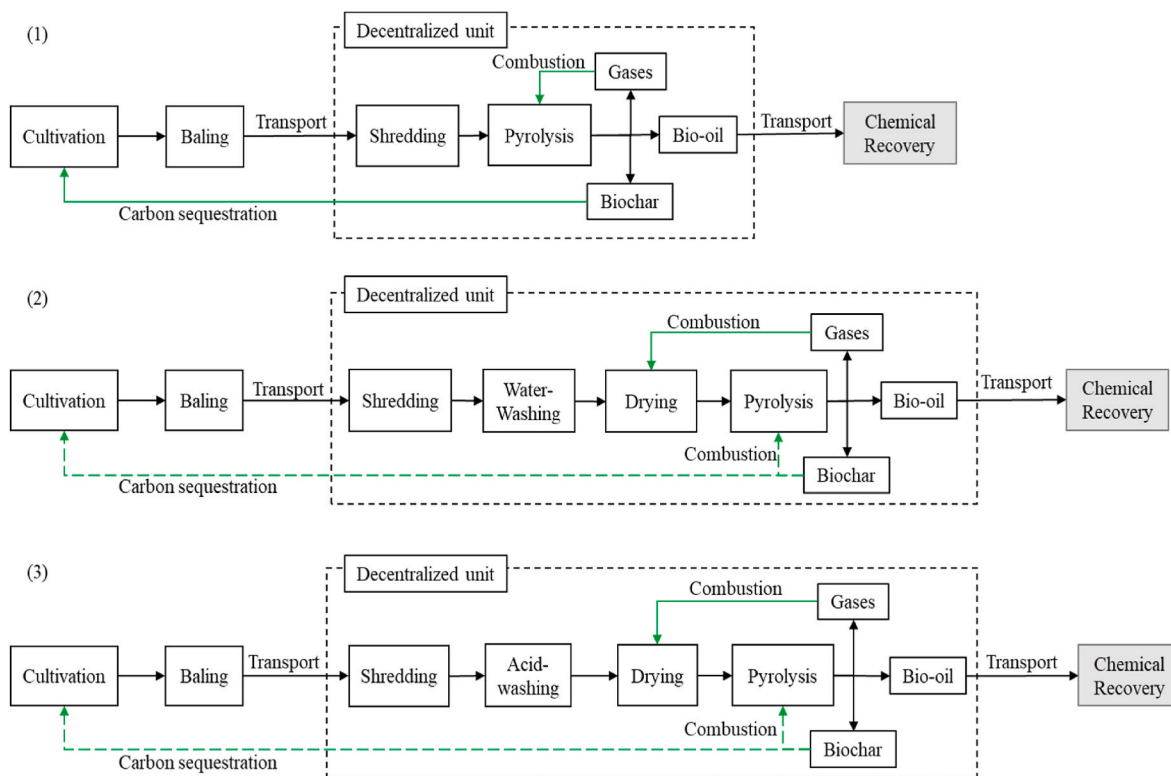


Fig. 2. Biorefineries with the pyrolysis of (1) unwashed straw; (2) water-washed straw; (3) acid-washed straw. The chemical recovery was not included in the system boundary.

loading-unloading operations are provided in the supporting information (Table S4 and Table S5).

2.3.2.4. Residue pretreatment.

a) Size reduction

Biomass obtained in bales was between 10 and 12 cm long, and its size was reduced using bale-shredders to 1–3 cm (longitudinal) before pyrolysis. The water content of straw at the time of harvest is <15% (ICAR, 2008), which means that it could be fed to the shredder without drying. The inventory for shredder operation (Supporting information, Table S6) was made based on the shredder efficiency of 81.7% (Sridhar and Suredrakumar, 2017).

b) Washing

As shown in Fig. 2, the biomass was also washed after shredding. The washing step removed a fraction of ash content in the biomass. For washing biomass with water, the straw to water ratio was maintained at 1:15 (w/w) for 60 min, followed by filtration and drying. The detailed washing procedure was reported previously by Bhatnagar et al. (2022). For acid-washing, bio-oil generated from pyrolysis of untreated straw was used as the acid because it comprised an aqueous phase rich in acetic acid (Bhatnagar et al., 2020) and could be generated at the pyrolysis unit. The straw to aqueous bio-oil (AqBO) ratio was maintained at 1:15 (w/w) for 60 min to wash the straw. The washed biomass was then neutralized with water before drying. The water to AqBO ratio was approximately 3:1 (w/w) for neutralization. The detailed acid-washing procedure was reported by Cen et al. (2019). The wastewater generated in both washing cases was not treated separately. It was assumed that wastewater could be discharged on the field since its pH (7.5–8.0) and conductivity (<4 dS/m) were within an acceptable range for irrigation water for straw crops (Bauder et al., 2014). The composition of untreated, water-washed, and acid-washed biomass is provided in the supporting information (Tables S7 and S8).

c) Drying

The wet biomass obtained from washing has 50% water, removed through drying at 60–80 °C. In this study, a fraction of biochar was burnt to supply heat for drying. The equations for calculating the energy required for drying were provided by Ding and Jiang (2013) and shown in the supplementary information. The specific heat value for RS was obtained from Dupont et al. (2014) and for WS from Chen et al. (2014). The actual heat required for drying was calculated by assuming heat losses of 15% (Brassard et al., 2018). The energy required for drying in each biorefinery case is given in the supporting information (Table S9) with the emission inventory.

2.3.2.5. Biomass pyrolysis. The experimental pyrolysis setup has been previously reported by Bhatnagar et al. (2020). Briefly, during two-step pyrolysis, the samples were heated from ambient temperature to 340 °C to obtain the first bio-oil fraction and from 340 °C to 600 °C in the second step. The heating rate during pyrolysis was 5 °C/min. The energy required for pyrolysis was obtained from the combustion of pyrolysis gases. The equations used in calculating the energy requirement is provided in supplementary information. The total energy for the pyrolysis was a sum of energy required to vaporize inherent moisture content and heat biomass to a pyrolysis temperature, enthalpy of pyrolysis and energy losses. The energy required for pyrolysis and the product yields is provided in the supporting information (Tables S7–S9).

2.3.2.6. Biochar application. As mentioned in Section 2.3.2.4, a fraction of biochar was used for biomass drying. But the residual char was used

for either carbon sequestration or energy production. The yield and composition of biochar are provided in the supporting information (Tables S10 and S11).

a) Soil amendment using biochar

Biochar was transported from the collection center through the same route as transporting bales and applied to the soil at 18 tons/ha (Mohammadi et al., 2016b). The fuel used for spreading biochar was assumed to be the same as baling, i.e., 4 dm³/ton char. Biochar from RS and WS was considered stable for >100 years (Bhatnagar et al., 2022), with up to 69% of its carbon content retained in the soil after 100 years (Mohammadi et al., 2016b). Although some estimates suggest that 80–90% of carbon from char was retained in the soil (G. Roberts K et al., 2009; Mohammadi et al., 2016a). The avoided CO_{2eq} (kg) were calculated using equation (1).

$$CO_{2eq} = C_{Char} C_r \left(\frac{44}{12} \right) \quad (1)$$

C_{Char} is the carbon content (kg) of biochar applied to the soil.

C_r is the carbon content (kg) remaining in the soil after 100 years.

44/12 is the factor for conversion of carbon to CO_{2eq}

Biochar also reduced NOx and methane emissions from soil (G. Roberts K et al., 2009). But due to uncertainty of measurement, this only implied that biochar did not create NOx or methane emissions. Biochar also reduces fertilizer requirements (Mohammadi et al., 2016b) by 20% for N-fertilizer and 50% for P and K-fertilizers. Due to uncertainty in measurements for the study area, it was assumed that no change in yield would occur. However, detailed meta-analysis report on biochar use in soil published by Joseph et al. (2021) showed that yields may increase between 10% and 42%, depending on soil types.

b) Biochar as a substitute for energy

Due to the similarity of biochar composition and Indian coal varieties (Table S11), biochar may be used in thermal power by substituting an equivalent amount of coal. The distance to the thermal powerplant from pyrolysis units was assumed to be the same distance as the chemical refinery because of an existing powerplant in Bathinda, Punjab. The emissions from char use in generating power are listed in the supporting information (Table S12). The electricity generated (in kWh) from biochar is given by equation (2).

$$Electricity\ generated = \frac{LCV_{Char} * M_{Char} * \eta_{electricity}}{100} \quad (2)$$

LCV_{Char} is the calorific value of biochar on the dry-ash-free basis in kWh/kg.

$\eta_{electricity}$ is the efficiency of thermal powerplants (30%)

M_{Char} is the amount of char in kg.

Another energetic biochar application was to substitute coal used for domestic purposes such as cooking and heating. It was assumed that biochar would be used in cookstoves locally, i.e., in the rural areas around the pyrolysis unit. The traditional cookstoves available have a thermal efficiency of 29.3%, while certain modified forced air draft cookstoves have a thermal efficiency of 36.56% (Kumar and Panwar, 2019). The emissions related to both traditional and modified cookstoves are listed in the supporting information (Table S13).

2.3.2.7. Transport to refinery. Bio-oil was transported in 6-ton trucks to the chemical refinery. The mileage for on-road trucks was assumed to be 8.5 km/dm³. In the case of direct transport of straw to the refinery, transport losses were assumed to be 0.5% by weight (Cardoen et al., 2015). The inventory is provided in the supporting information (Table S14).

2.3.3. Life cycle impact assessment

The current study evaluated the global warming potential (GWP) of pyrolysis using the IPCC 2021 methodology (100-year perspective). Sensitivity analysis was performed by varying key inputs by $\pm 20\%$ and reevaluating the GWP of pyrolysis for unwashed RS and WS.

2.3.4. Economic assessment

The economic assessment of the pyrolysis process was based on 1-ton biomass pyrolyzed. The equipment costs were obtained from online retailers for industrial goods and are listed in the supporting information. The operational expenses were calculated based on assumptions provided in *Ag Decision Maker* (2015). The freight transport and diesel costs were obtained from an online repository of statistical surveys called IndiaStat. All the expenses involved are listed in the supporting information (Tables S15 and S16).

The current study only discusses the minimum product selling price (in USD/ton) for biochar ($BC - 0$), calculated from equation (3). Additional revenue may be derived using equation (4) from carbon credits gained by avoiding $\text{CO}_{2\text{eq}}$ emissions (Cheng et al., 2020). Four carbon price benchmarks have been proposed—€10, €30, €60, and €120 based on European markets (OECD, 2021) for every avoided ton of $\text{CO}_{2\text{eq}}$.

$$BC - 0 = \frac{\text{Sum of costs incurred over plant lifetime}}{\text{Sum of biochar produced over plant lifetime}}$$

$$BC - 0 = \frac{\text{Capex}_{\text{total}} + \sum_{t=1}^n (\text{OM}_t + L_t + F_t - H_t) / (1 + r)^t}{\sum_{t=1}^n \text{Char}_t / (1 + r)^t} \quad (3)$$

OM_t is the annual operation and maintenance cost.

L_t is the annual labor cost.

F_t is the annual cost of feedstock production.

H_t is the annual cost of heat substitution (assumed 0 in this study)

r is the discount rate (10.7% in this study)

t is the lifetime of a plant in years (1, 2, ..., n)

n is total plant life in years (20 years in this study)

$$BC - i = \frac{BC - 0 + \text{GWP} \left(\frac{\text{tCO}_{2\text{-eq}}}{\text{tfeedstock}} \right)}{Y_{\text{char}}} \times \text{Carbon credit} \left(\frac{\text{€}}{\text{tCO}_{2\text{-eq}}} \right) \times \text{Exchange rate} (\text{€ to USD}) \quad (4)$$

$BC - i$ is biochar value for various carbon price benchmarks (10, 30, 60, 120 €/t $\text{CO}_{2\text{eq}}$)

$\text{GWP} \left(\frac{\text{tCO}_{2\text{-eq}}}{\text{tfeedstock}} \right)$ is the total emissions from the biorefinery (without product substitution)

Y_{char} is the yield of char in kg/kg-straw.

3. Results and discussions

3.1. Pyrolysis product yields and energy requirements

The bio-oil yield from all pyrolysis cases for RS ranged from 25 to 34% and for WS from 31 to 41%. After washing, the ash content of RS was reduced because the inorganic elements in the biomass are found as water-soluble and acid-soluble ions and on mixing biomass with water or AqBO, these ions are leached out (Cen et al., 2019). Details about the role of bio-oil composition, i.e., presence of acetic acid and phenolics, in the acid-washing of biomass have been published by Chen et al. (2021, 2020). Specifically, sodium, potassium, calcium, and magnesium promote cracking of bio-oil vapors to yield more pyrolysis gases and biochar than bio-oil. Therefore, the bio-oil yields increased by 4% after water-washing and 9% after acid-washing the biomass. Reportedly, acid-washing RS also improved the relative yield of levoglucosan from 2.1% for the unwashed sample to 31.2% (Cen et al., 2019). A similar increase in yields of levoglucosan was observed from WS. Since levoglucosan is a high-value chemical with potential uses in synthesis of

biodegradable polymer, antiviral agents, and propellants (Rover et al., 2019), it is important to maximize its yield in bio-oil. Washing the biomass also selectively increased the yield of polyphenols (antioxidants and flavoring agents) over monophenols in the bio-oil (Zhang et al., 2017). Hence, washing the biomass is an important step, which may lead to production of a bio-oil that has a higher commercial value than bio-oil generated without biomass washing.

The energy required for biomass drying (after washing) was obtained from biochar combustion. Between 37% and 50% of the biochar generated during pyrolysis was used for drying. The energy required for the pyrolysis step was entirely met by pyrolysis gas combustion. These results suggest that it is possible to set up a pyrolysis unit that does not require an external heat supply, making it self-sustainable.

However, if biochar from washed straw is used for drying, less of it would be available for carbon sequestration. Therefore, there would likely be a trade-off between improving chemical yields and the environmental performance of the decentralized biorefinery, as discussed later in section 3.2.3.

3.2. Lifecycle global warming impacts of biorefinery

3.2.1. Impact of cultivation

All cultivation activities require electricity or diesel for running the equipment. The current study allocated the impact of the cultivation between grain and straw based on their economic value. This allocation was contrary to many studies (Singh and Basak, 2019; Soam et al., 2017; Sree Kumar et al., 2020), where system boundary starts from residue collection and considers residue to be free from the impact of the cultivation practices.

As shown in the supporting information (Fig. S1), the net GWP for RS is higher than for WS cultivation. This difference is attributed to rice's more energy-intensive cultivation practices than wheat. Rice has a higher land, seed, fertilizer, manure, pesticides, and irrigation requirement per ton of straw produced.

The agricultural activities listed in Table S2 are classified into—land preparation (irrigation and land-use), energy (diesel and electricity used for tillage, spreading, harvesting), fertilizers (production and transport), and pesticides (production and transport). Soni et al. (2018) have identified fertilizer application with tractor-operated spreaders, increased mechanization on large farms, and high pumping requirements for irrigation (especially rice) are the key drivers of emissions in the rice-wheat cultivating system in India. Therefore, GWP related to cultivation is not likely to change unless overall farming scenarios evolve.

3.2.2. Impact of transport, pretreatment, and pyrolysis

The net GWP of decentralized biorefineries is presented in Fig. 3. This figure does not include the avoided emissions from open burning or avoided $\text{CO}_{2\text{eq}}$ from biochar use. When cultivation is included, RS-based biorefinery has a higher GWP than WS-based. However, the GWP of biorefineries from both straws is similar without the cultivation step. The GWP for unwashed straw was 54–55 kg $\text{CO}_{2\text{eq}}$ /ton and washed straw was 59–61 kg $\text{CO}_{2\text{eq}}$ /ton.

In the absence of cultivation, the biggest contribution to GWP was the size reduction step, wherein grid-connected electricity was used. Since coal-based thermal power plants supply 66% of the electricity in Punjab, the GWP of this step was the highest.

The next major contribution to GWP was from the transportation of materials. The short-distance transport includes transport of biomass from farm to collection center and transport of char back to farm for soil application. It was reported by Bhatnagar et al. (2022) that biomass washing leads to a mass loss of up to 11%. Hence, more biomass needs to be recovered from the field to meet the required 1-ton dried straw input for pyrolysis, which would add to the payload transported. However, surplus biochar transported to farms reduces when biomass is washed because biochar is consumed in drying the biomass after washing. The

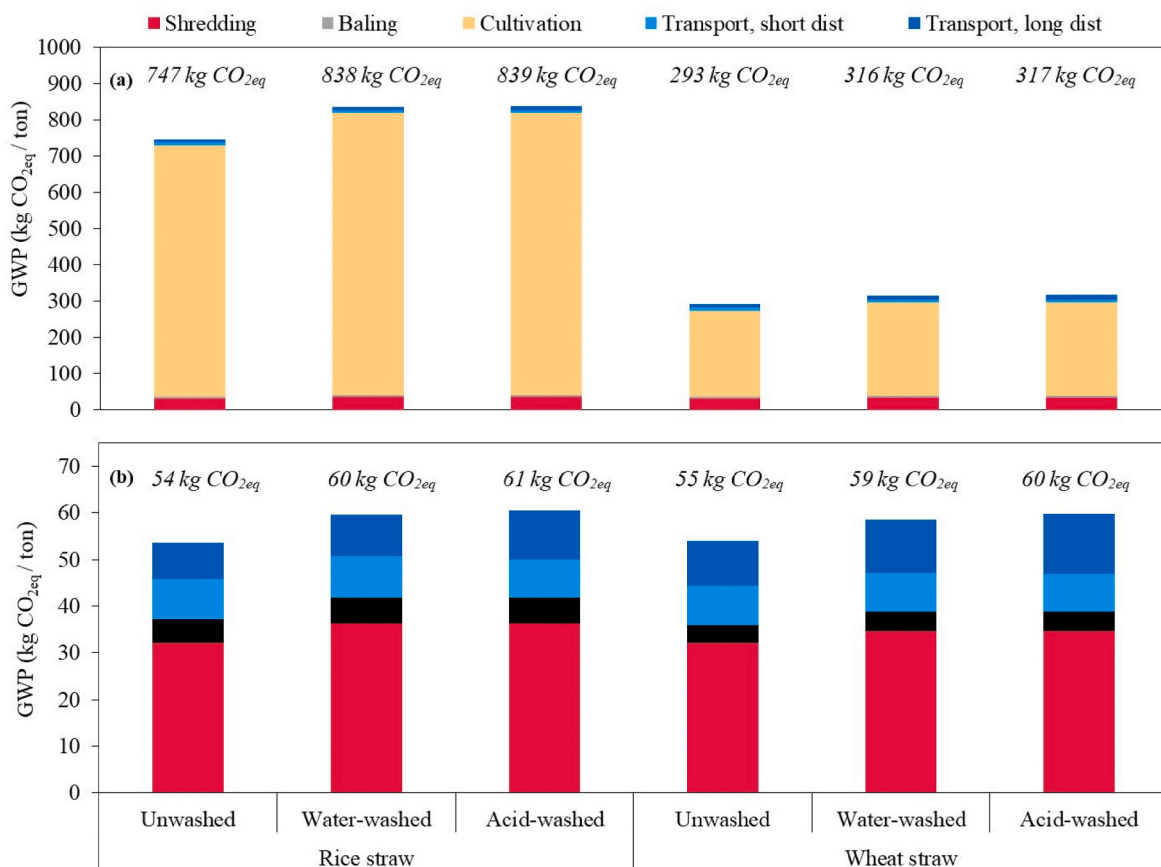


Fig. 3. Comparison of GWP (kg CO_{2eq}) for each pyrolysis case (a) with cultivation step and (b) without cultivation.

total change in GWP is then dependent on the balance of biomass and biochar transport. Long-distance transport refers to the transport of bio-oil to the chemical refinery for further processing. As the bio-oil yield increased, the GWP of this step increased. For the transport of bio-oil from the water-washed straw, the GWP was 12.5–22.2% higher than unwashed straw, and for acid-washed straw, it was 25–44% higher.

3.2.3. Impact of biochar use in soil

The environmental performance of biorefinery when biochar was used in the soil is shown in Fig. 4. The GWP credits, shown as negative

values, included avoided emissions from open burning, avoided chemical fertilizer (N–P–K), and avoided CO_{2eq} from putting char in soil. Due to uncertainty in measurement, it is assumed that biochar from all pyrolysis cases substituted the same amount of fertilizers.

Several key observations were made from the results presented in Fig. 4. First, biochar from WS acted as a better carbon sink than biochar from RS, although more biochar was obtained from RS (27–38%) pyrolysis than WS (24–34%). This improved performance from WS was due to its higher carbon content (65%), shown in Table S17.

Second, avoided emissions from open burning of RS (–286 kg CO_{2eq})

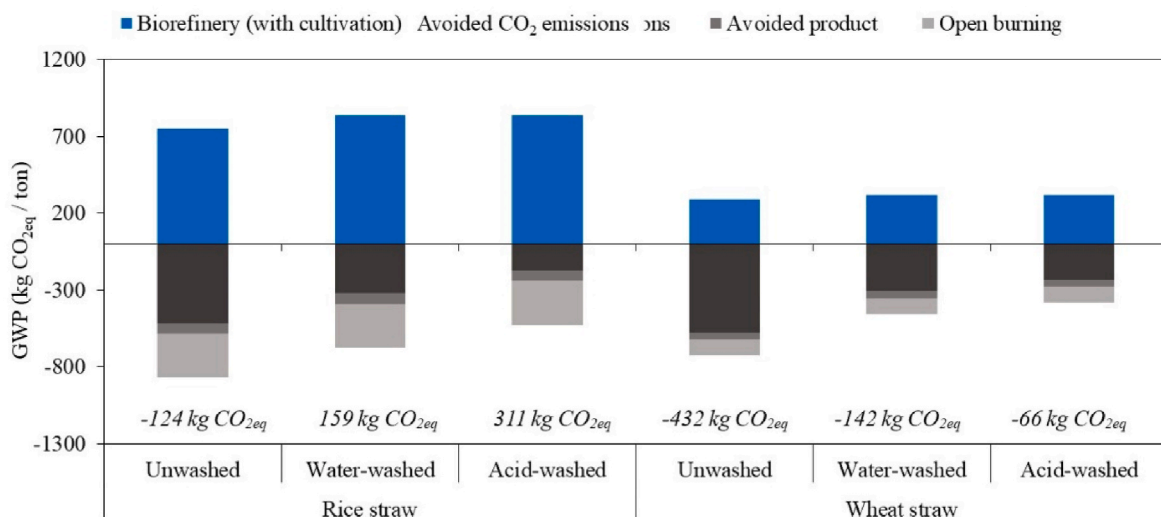


Fig. 4. Global warming impact of biorefineries (with cultivation) when biochar was applied to the soil.

were higher than WS (-106 kg CO_{2eq}). This difference was due to the higher ash content of RS than WS, which reduces the combustion efficiency of biomass, releasing more methane, CO, and particulates during open-field burning (Sahu et al., 2021), shown in supporting information (Table S18).

Third, the unwashed pyrolysis had the lowest net GWP for RS and WS-based biorefineries. Washing led to a net positive GWP value for RS-based biorefinery, which was undesirable. The difference in pyrolysis of unwashed and washed biomass was due to the biochar carbon content and total char availability after biomass drying. Although the carbon content of biochar continuously increased from washing (Table S17), its use in drying led to reduced availability.

It is possible to replace biochar-based boilers for biomass drying with solar thermal dryers or combusting dry straw to provide the heat requirements. Then, GWP for water-washed straw pyrolysis would reduce by ~50% and acid-washed pyrolysis by ~40% due to more char available for sequestration. Additionally, if higher C_r values (80–90%) were assumed, the avoided CO₂ emissions from biochar of water-washed straw were higher than for unwashed straw due to similar total yield. However, solar drying is slower than conventional methods and is subject to seasonal changes (Udomkun et al., 2020) and straw has a propensity to form low-melting ash that may damage boilers faster (Bhatnagar et al., 2022). Therefore, if only surplus char were available for use, unwashed pyrolysis was the preferred route for straw management.

3.2.4. Impact of alternative biochar applications: power and heat

Fig. 5 showed the net GWP for biorefineries when biochar was used in soil or substituted coal in thermal power and cookstove. When biochar was used in thermal power, the avoided product was electricity generated by coal, and in cookstoves, it was the heat generated from burning coal.

It was evident that substituting coal with biochar for power generation offers the most benefits due to the lower emission factors for biochar-based power production compared to coal (Soam et al., 2017). Peters et al. (2015a) also observed that the most favorable biochar application was substituting fossil coal. Even assuming C_r of 90% for biochar from unwashed RS the net GWP (-275 kg CO_{2eq}) for its use in soil, was lower than GWP for biochar use in power generation (-646 kg CO_{2eq}). Although for WS-derived biochar, at a C_r of 90%, biochar use in soil was more favorable than in power plants. Biochar used in cookstoves for heat generation as a substitute for coal did not offer similar benefits as its use in power plant, but it was better than biochar use in soil. However, there is a governmental push through policy changes to substitute these solid-fuel-based cookstoves with cleaner gaseous fuels

(Sharma and Jain, 2019); hence, using biochar for heating would not be sustainable.

It should be noted that considering all biochar-derived CO₂ as biogenic may lead to underestimating the net GWP due to the uncertainty in time frames and efficiency of biomass uptake of the released CO₂ (Matušítk et al., 2022).

3.3. Decentralized vs. centralized biorefinery

The direct biomass transfer to the refinery always performed worse than decentralized pyrolysis of biomass, as shown in Table 1, due to the transport logistics involved. The long-distance transport comprises two parts, sending the biomass from the farm to a chemical refinery for pyrolysis and transporting biochar back to the farm for application to a field or use in domestic heating. In direct biomass transfer, for 1-ton of residue pyrolyzed, 1.65-ton RS was transported as bales, which increased the transport payload compared to only bio-oil transport (0.25–0.3 ton), increasing the GWP of the transport step by 8 times.

Further, in all biochar applications, the trend in GWP savings was similar to decentralized treatment, and biochar in power generation was the best application for both RS (-615 kg CO_{2eq}/ton) and WS (-846 kg CO_{2eq}/ton). This trend indicated that substituting the fossil-based emissions with the biochar-based alternative was the best way to reduce the GWP of a biorefinery.

Based on the annual operation of the decentralized biorefineries operating in the ten selected districts of Punjab (Fig. 1), potentially

Table 1
Process-wise distribution of GWP for direct transfer of biomass to a centralized pyrolysis facility.

Unit (kg CO _{2eq} /ton straw)	Char in soil (C _r : 69%)		Char in powerplant		Char in cookstove	
	RS	WS	RS	WS	RS	WS
Net GWP	-75	-384	-615	-846	-166.6	-414.6
Biorefinery impact	-518	-577	8	8	0	0
Shredding	32	32	32	32	32	32
Baling	4.9	3.7	4.9	3.7	4.9	3.7
Cultivation	690	239	690	239	693.2	238.6
Transport, short distance	8.8	8.6	7.1	7.1	7.1	7.1
Transport, long distance	63	61	51	51	63	63
Char spread	0.8	0.7	0.0	0.0	0.0	0.0
Avoided product	-69	-47	-1122	-1080	-681	-655
Open burning	-286	-106	-286	-106	-286	-106

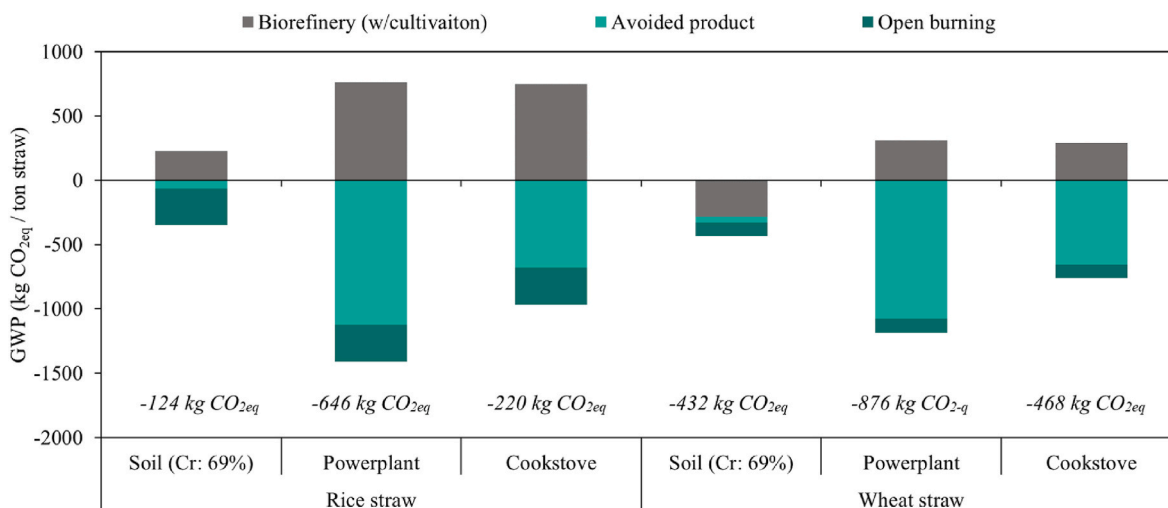


Fig. 5. GWP (kg CO_{2eq}/ton) of biorefineries for various char applications.

12.55-kt of biochar from RS and 11.50-kt biochar from WS may be available for use. Using straw for pyrolysis units yields an annual saving of 9.36 Gg CO_{2eq} from avoided RS burning and 3.5 Gg CO_{2eq} from WS burning. The biochar may be applied to 639–697 ha of soil at 18 tons/ha, allowing the sequestration of 19–20 Gg CO_{2eq}. Whereas biochar use in thermal power avoided 35–36 Gg CO_{2eq} emissions annually.

Other studies have reported crop residue use through thermochemical and biological processes, apart from pyrolysis. Sreekumar et al. (2020) evaluated bioethanol production from rice straw and reported a net GWP of −0.392 kg CO_{2eq}/liter ethanol, i.e., about −116 kg CO_{2eq}/ton straw. Soam et al. (2017) reported that the GWP of four applications of rice straw was in the order: field incorporation (1025 kg CO_{2eq}/ton straw) > animal fodder (−185 kg CO_{2eq}/ton straw) > biogas production (−1023 kg CO_{2eq}/ton straw) > electricity (−1471 kg CO_{2eq}/ton straw). Finally, Singh and Basak (2019) compared the GWP of thermochemical and biological processes using rice straw. They reported a net GWP of −1220 kg CO_{2eq}/ton straw by incineration and −1343 kg CO_{2eq}/ton straw by gasification. Anaerobic digestion and fermentation have a net GWP of −1162 kg CO_{2eq}/ton straw and −152 kg CO_{2eq}/ton straw. Thermochemical conversion routes performed better because of the electricity generation potential, similar to this study's results for pyrolysis. When comparing these results with the present study, it should be noted that the other studies assumed no impact of cultivation on straw management.

3.4. Economic assessment of biorefinery

Another aim of the current study was to propose a selling price for the biochar generated from straw and compare its competitiveness with commercially available biochar sold in the Indian market. The current range of biochar prices charged by commercial retailers in India is USD 234–1300/ton char, based on the difference in char quality.

Fig. 6 shows the projected biochar selling price with and without including the benefits of a carbon pricing mechanism. Because the biochar yield is highest from unwashed straw, it has the lowest selling price. Since the price of biochar, in this case, was calculated without accounting for market mechanisms that determine the bio-oil selling price, these values may be subject to change. Further, there is no standardized quality of biochar at present, and with standardization of product quality, further expenses may be incurred, leading to a change in selling price.

3.5. Sensitivity analysis

According to the ISO standards for LCA, a sensitivity analysis is vital in evaluating the role of each parameter towards the overall environmental impact of the product/process analyzed. Hence, for the sensitivity analysis, the inventory parameters and expenses involved in setting up the biorefinery were changed one at a time by a factor of ±20%. The results were reported for the pyrolysis of unwashed RS without product substitution.

The input parameters investigated for GWP results were change in straw yield, baler collection efficiency, the distance of collection center from the field, shredding efficiency, bio-oil yield from pyrolysis, and the distance from collection center to the refinery for chemical recovery from bio-oil. The analysis results (Fig. 7a) show that GWP values were not sensitive to the bio-oil yield and transport distances. A variation (±20%) of these parameters led to a <1% change of GWP from base values. Therefore, even if the refinery location had a certain degree of uncertainty, it would have a relatively small influence on the LCA. The other three parameters caused much larger changes in GWP of base case values, and the sensitivity was in the order shredding efficiency > baler efficiency > straw yield.

The shredder efficiency (81.7%) was related to the electricity demand at a collection center. An improvement in efficiency reduced the operational hours of the shredder, which reduced the requirement for grid-connected electricity that mainly comes from thermal power. It also reduced the amount of biomass required from the field for obtaining the functional unit of 1-ton size reduced straw. When the shredder efficiency increased, the net GWP reduced by 30%, and when the efficiency reduced, the net GWP increased by 5%. This result was in contrast to Winjobi et al. (2016), who found did not find size reduction to be a sensitive parameter in biomass pyrolysis, possibly due to different choice of the functional unit.

Similar to shredding, the baler efficiency (74.6%) determined the operational hours and the diesel required for collecting the required biomass. An improvement in collection efficiency (+20%) reduced the net GWP by 16%, while a reduction in collection efficiency (−20%) increased the GWP by 24%. These results suggest that technology improvements could effectively lower the operational hours of equipment and reduce the environmental impacts of a process. This dependence on technology was also observed by other studies (Sahoo et al., 2021; Zheng et al., 2018). Finally, cultivation in India is heavily dependent on the monsoon season (Directorate of Economics and Statistics, 2019), which implies straw availability may change annually. But uncertainty in straw availability influenced GWP to a lesser extent (5–10% of base values)

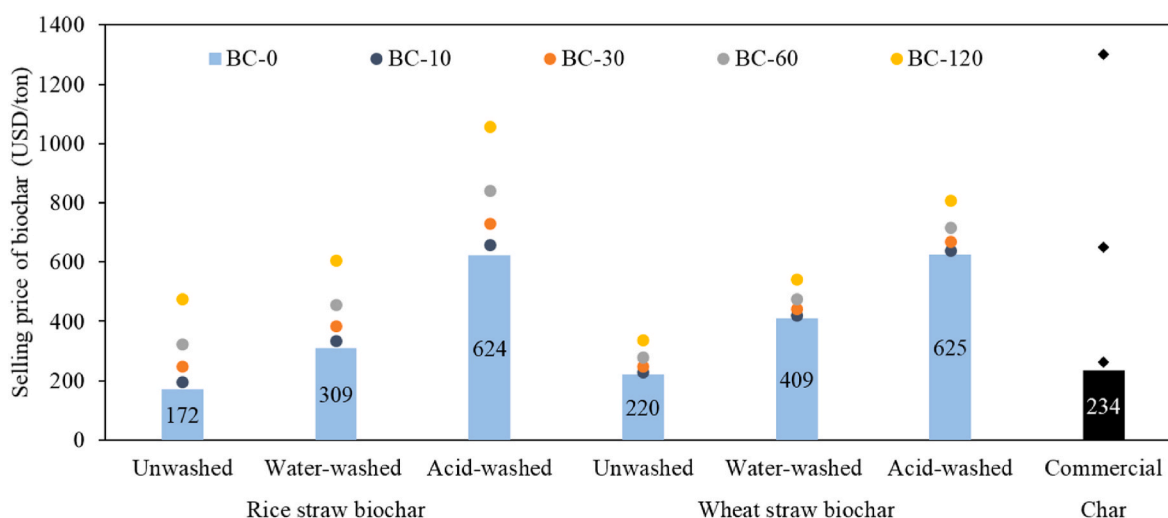


Fig. 6. Projected selling price of biochar with and without the carbon price mechanism (BC-0,10,30,60,120 are prices as per carbon price benchmarks).

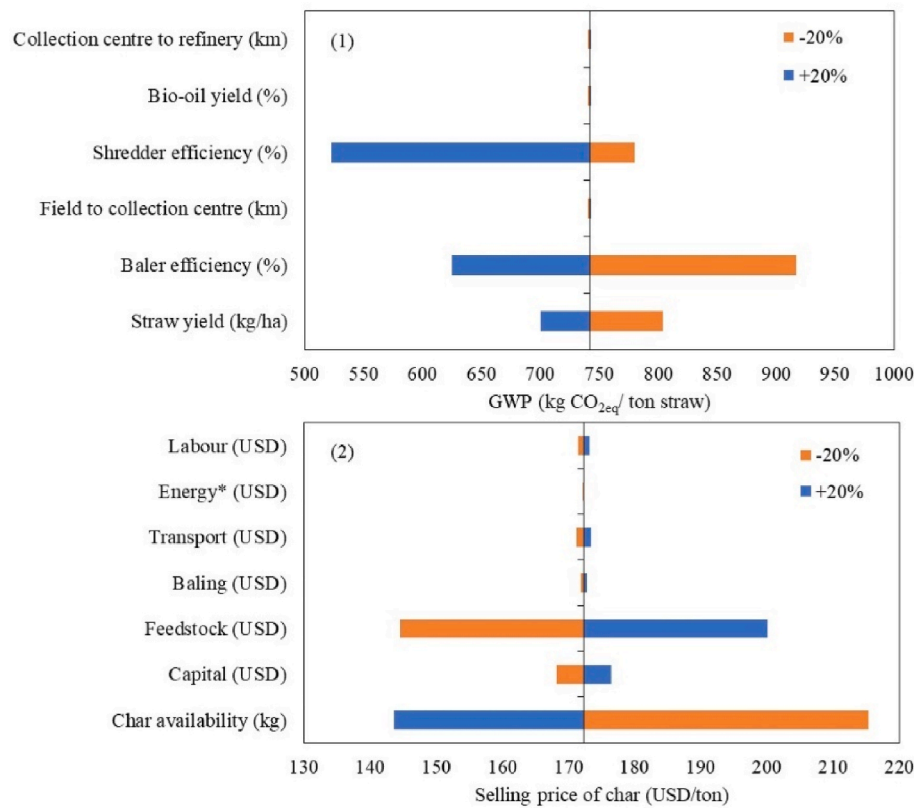


Fig. 7. Sensitivity analysis (1) the environmental impact of, and (2) biochar selling price (*Diesel and electricity use) for the pyrolysis of unwashed rice straw without product substitution.

than the technological barriers.

A sensitivity analysis of the minimum selling price of biochar included annual costs incurred for the purchase of straw from the farm, collection of straw through baling, capital, labor, energy (electricity and diesel) and transport, and annual biochar availability. The results of the analysis (Fig. 7b) show that the price of biochar was most sensitive to changes in the annual availability of char and the purchase price of RS (INR 2.46/kg). The feedstock price depends on its competitive use as a food source during animal husbandry (Duncan et al., 2020). Biochar's selling price was marginally (<2.5% change from base values) sensitive to capital and not sensitive (<1% change from base values) to changing costs of energy or transport within the fluctuation factor of $\pm 20\%$ used in the current study.

Notably, the current study followed a conservative approach when quantifying GWP and the minimum selling price of char to avoid overestimating the benefits of a biorefinery approach for residue management.

4. Conclusion

The current study showed that decentralized pyrolysis of unwashed straw is an environmentally more suitable alternative for crop residue management. However, washed straw produces more high-value chemicals in the bio-oil than unwashed straw. The decentralized biorefinery proposed in this study offers environmental savings from avoided emissions through open burning of RS (-285 kg CO_{2eq}/ton straw) and WS (-106 kg CO_{2eq}/ton straw) on the field and from using biochar as a carbon sink in soil. The pyrolysis unit could be self-sustaining using pyrolysis gases and biochar. The net GWP from decentralized pyrolysis of RS was -124 kg CO_{2eq}/ton straw compared to -75 kg CO_{2eq}/ton straw for centralized biorefinery, indicating that it was better to treat biomass locally. Further, with an all-year-round operation, biochar could be sold at 172–625 USD/ton, which was

competitive with current market prices. Therefore, biorefinery based on two-step slow pyrolysis of rice or wheat straw proposed in this study is techno-economically and environmentally competitive.

CRediT authorship contribution statement

Anubhuti Bhatnagar: Conceptualization, Investigation, Methodology, Validation, Writing – original draft. **Poonam Khatri:** Conceptualization, Investigation, Methodology, Validation, Writing – review & editing. **Malgorzata Krzywonos:** Methodology, Validation, Writing – review & editing. **Henrik Tolvanen:** Supervision, Writing – review & editing. **Jukka Konttinen:** Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2022.132998>.

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