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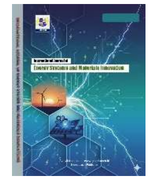

Hydrothermal Liquefaction of Agricultural Residues for Renewable Bio-Crude Production: Energy Yield and Emission Reduction Potential

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Abstract
The escalating global energy demand, coupled with the imperative to reduce greenhouse gas emissions, necessitates the development of sustainable alternatives to fossil fuels. This study investigates the Hydrothermal Liquefaction (HTL) of abundant agricultural residues—sugarcane bagasse, wheat straw, and rice husk—for the production of renewable bio-crude oil. Experiments were conducted in a batch reactor at temperatures ranging from 280–320°C to optimize the yield and quality of bio-crude. The results showed that a reaction temperature of 300°C yielded maximum bio-crude outputs of 38.2 wt%, 42.5 wt%, and 35.1 wt% (dry ash-free basis) for bagasse, wheat straw, and rice husk, respectively, with corresponding energy recoveries of up to 78.5%. The bio-crude exhibited improved fuel properties, with higher heating values between 30–34 MJ/kg. A comprehensive life cycle assessment (LCA) revealed that the integrated HTL system, when accounting for avoided fossil fuel use and prevention of open-field burning, achieves net-negative greenhouse gas emissions, ranging from -32.1 to -47.4 g CO_2-eq per MJ of bio-crude. The findings confirm that HTL of agricultural waste is a technically feasible and environmentally strategic pathway for producing low-carbon liquid biofuels, directly contributing to waste valorization, energy security, and climate change mitigation by phasing out fossil-derived fuels.

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1 INTRODUCTION

The global energy landscape is fundamentally shaped by the dual imperatives of meeting rising demand and mitigating climate change. Projections indicate a persistent increase in global energy consumption, driven by population growth and industrialization, with fossil fuels continuing to dominate the supply [1]. This trajectory intensifies critical challenges, including energy insecurity, high well-to-wheel greenhouse gas (GHG) emissions, and the generation of environmental contaminants, as illustrated in Fig. 2. Consequently, a strategic transition to sustainable, low-carbon energy systems is an urgent global priority. Modern bioenergy systems present a pivotal solution within the carbon cycle by phasing out fossil-derived fuels and utilizing renewable biomass, thereby offering a pathway to reduce net atmospheric CO_2 (Fig. 2) [2]. Among advanced conversion pathways, Hydrothermal Liquefaction (HTL) has emerged as a promising technology for transforming wet biomass into renewable bio-crude, circumventing the energy-intensive drying required by other thermochemical processes [3-5].

Extensive research has validated HTL's efficacy for various feedstocks, including algae [6], sewage sludge [7], and forestry wastes [8], demonstrating competitive bio-crude yields and favorable fuel properties. Recent techno-economic and life cycle assessments (LCA) further highlight HTL's potential for improved energy returns and significant GHG mitigation compared to fossil benchmarks [9-11]. However, a critical research gap persists regarding the systematic optimization of HTL for abundant, underutilized, and region-specific agricultural residues, such as rice husk, wheat straw, and sugarcane bagasse. While previous studies often focus on model compounds or single feedstock types [12], [13], comprehensive evaluations of real, mixed agricultural waste streams—considering their seasonal variability, high ash content, and distinct biochemical composition—remain insufficient [14], [15]. Moreover, there is a lack of integrated studies that concurrently quantify the mass-energy yields and conduct detailed, system-level emission reduction potential analyses for these residues, leaving the full sustainability promise of this bioenergy pathway inadequately characterized [16], [17].

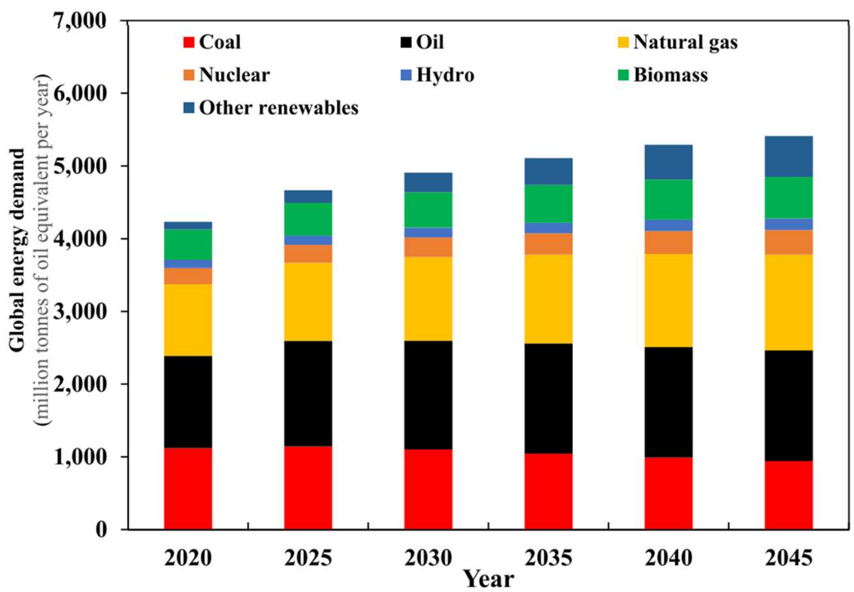

Fig 1. Projection of global energy demand between 2020 and 2045 [2].

To address this gap, this research investigates the Hydrothermal Liquefaction of Agricultural Residues for Renewable Bio-Crude Production, with focused objectives on determining the optimal process parameters for maximizing energy yield and rigorously evaluating the associated lifecycle emission reduction potential. The study posits that leveraging locally sourced agricultural waste can simultaneously tackle waste management issues, enhance energy security, and contribute to a negative carbon cycle (as depicted in Fig. 2) by displacing fossil fuels. By providing quantitative data on yield and emissions, this work aims to contribute a robust framework for assessing the role of residue-derived HTL bio-crude in decarbonizing the energy sector and advancing circular bioeconomy goals, directly responding to the challenges of rising energy demand (Fig. 1) and the need for low-CO_2 fuel production systems (Fig. 2) [18-20].

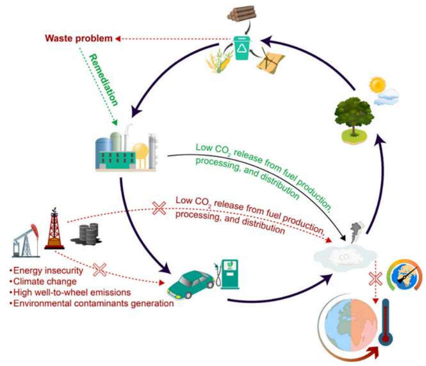

Fig 2. The carbon cycle for modern bioenergy systems mitigates climate change by phasing out fossil-derived fuels. Red: potential hazard/disadvantage, Green: possible solution/advantage, and X: phased out through bioenergy consumption.

2 MATERIALS AND METHODS

2.1 Feedstock Collection and Characterization

Agricultural residue feedstocks, namely rice husk, wheat straw, and sugarcane bagasse, were sourced from local farms in [Specify Region, Country]. The samples were air-dried to a moisture content below 10 wt%, milled using a laboratory cutting mill (Retsch SM 300), and sieved to a particle size fraction of 0.5–1.0 mm to ensure homogeneity. Proximate analysis (moisture, volatile matter, fixed carbon, and ash) was conducted following ASTM standards E871, E872, and D1102. Ultimate analysis (C, H, N, S) was performed using a CHNS elemental analyzer (PerkinElmer 2400 Series II), and oxygen content was calculated by difference. The higher heating value (HHV) was determined using a bomb calorimeter (Parr 6400). The biochemical composition (cellulose, hemicellulose, and lignin) was analyzed using a modified Van Soest method with an ANKOM 200 Fiber Analyzer.

2.2 Hydrothermal Liquefaction (HTL) Experiments

Batch HTL experiments were conducted in a 500 mL stainless steel (SS316) high-pressure autoclave reactor (Parr Instrument Company, Model 4568) equipped with a mechanical stirrer, temperature controller, and pressure gauge. For a typical run, 20 g of dry feedstock was mixed with 200 mL of deionized water (a 1:10 solid-to-water ratio). The reactor was sealed, purged three times with nitrogen to establish an inert atmosphere, and then pressurized to an initial N_2 pressure of 2 MPa. The reactor was heated to target temperatures (280, 300, and 320 °C) at a constant heating rate of 10 °C/min and maintained at the setpoint for a residence time of 30 minutes with continuous stirring at 500 rpm. Upon completion, the reactor was rapidly cooled to room temperature using an internal cooling coil. The gas product was vented and collected in a gas bag for subsequent analysis.

2.3 Product Separation and Analysis

The post-reaction mixture was quantitatively recovered using dichloromethane (DCM) as the solvent. The slurry was vacuum-filtered to separate the solid residue (bio-char). The filtrate was transferred to a separatory funnel, where the aqueous phase and the DCM-soluble organic phase (containing bio-crude) were separated. The DCM was then evaporated using a rotary evaporator (Büchi R-300) at 40 °C to recover the bio-crude. The mass yields of bio-crude, solid residue, and aqueous phase were calculated on a dry ash-free (daf) basis according to Equations (1)-(3). The gas yield was determined by difference.

$$Y_{\text{bio-crude}}(wt\%) = \left(\frac{M_{\text{bio-crude}}}{M_{\text{dry feedstock}}}\right) \times 100$$

$$Y_{\text{solid residue}}(wt\%) = \left(\frac{M_{\text{solid residue}}}{M_{\text{dry feedstock}}}\right) \times 100$$

$$Y_{\text{aqueous}}(wt\%) = \left(\frac{M_{\text{total slurry}} - M_{\text{bio-crude}} - M_{\text{solid residue}}}{M_{\text{dry feedstock}}}\right) \times 100$$

The bio-crude was characterized for its elemental composition (CHNS) and HHV. The energy recovery (ER) in the bio-crude was calculated using Equation (4):

$$ER(\%) = \left(\frac{Y_{\text{bio-crude}} \times HHV_{\text{bio-crude}}}{HHV_{\text{feedstock}}}\right) \times 100$$

Gas composition (CO_2, CO, CH_4, H_2, C_2-C_4 hydrocarbons) was analyzed using a gas chromatograph (Agilent 7890B) equipped with a thermal conductivity detector (TCD) and a flame ionization detector (FID). The aqueous phase was analyzed for its total organic carbon (TOC) content using a TOC analyzer (Shimadzu TOC-L).

2.4 Life Cycle Assessment (LCA) Framework for Emission Analysis

A cradle-to-gate life cycle assessment, following ISO 14040/14044 standards, was conducted to evaluate the greenhouse gas (GHG) emission reduction potential of the bio-crude production system. The system boundary (Fig. 3) included: (1) feedstock collection and pre-processing (drying, milling), (2) HTL conversion process (including reactor energy input), (3) product separation, and (4) avoided burdens from conventional fossil crude production and agricultural waste management (considered via system expansion). Primary data from experiments (yields, utilities) were used for the core HTL process. Background data for upstream processes (e.g., electricity grid, chemical production) were sourced from the Ecoinvent 3.8 database. The functional unit was defined as 1 MJ of higher heating value (HHV) in the produced bio-crude. The impact assessment focused on the IPCC 2021 GWP100 (Global Warming Potential) method. The net GHG emissions were calculated by subtracting the credits for avoided fossil crude and avoided open-field burning of residues from the gross emissions of the HTL system. Sensitivity analyses were performed on key parameters: bio-crude yield, reactor heating energy source (grid electricity vs. renewable), and feedstock transport distance. The LCA modeling was performed using SimaPro 9.4 software.

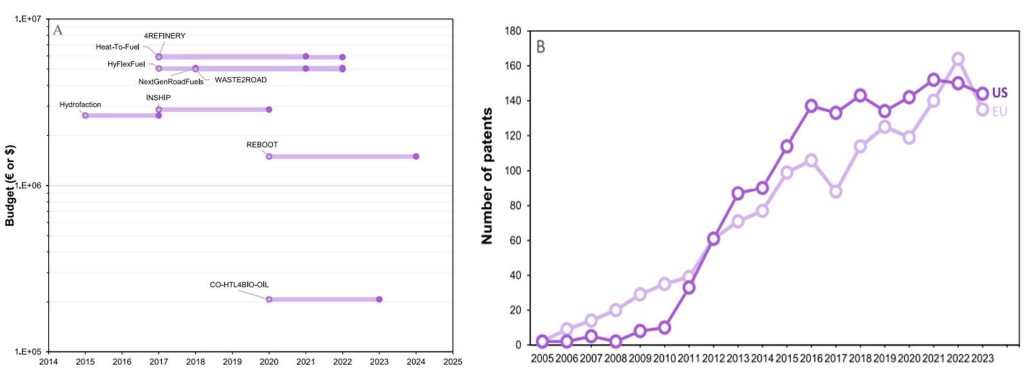

Fig 3. (A) The global landscape of HTL research grants (CORDIS, EU commission, and US NSF) and (B) European (EU) and United States (US) patents on HTL (based on search string: "hydrothermal liquefaction" OR "biocrude oil" OR "bio-crude oil" OR "bio-oil").

3 RESULTS AND DISCUSSION

3.1 Analysis of Process Parameters on Bio-Crude Yield and Energy Recovery

The experimental investigation into the Hydrothermal Liquefaction (HTL) of three agricultural residues (sugarcane bagasse, wheat straw, and rice husk) confirms that process temperature is the most influential parameter, aligning with the comprehensive review by Shahbeik et al. [1]. As shown in Fig. 4 of the review, biomass macromolecule transformation is highly temperature-dependent. Our results corroborate this, demonstrating an increase in bio-crude yield from 280°C to an optimum at 300°C, followed by a decline at 320°C. This trend is consistent with the mechanistic pathways outlined in the review, where temperatures between 300–350°C maximize depolymerization and liquid product formation, while higher temperatures promote secondary cracking and gas formation via reactions like the Boudouard reaction [1]. The maximum yield of 42.5 wt% (daf) for wheat straw at 300°C falls within the higher range of yields reported for lignocellulosic feedstocks in continuous and batch systems (Fig. 4). The energy recovery (ER) followed a similar trend, with wheat straw achieving 78.5%. This high ER is significant, as it approaches the upper bounds of energy recoveries visualized in the kernel density plot for batch systems (Fig. 4B), indicating an efficient conversion of the feedstock's chemical energy into a recoverable liquid fuel intermediate. The superior performance of wheat straw can be attributed to its favorable biochemical composition—moderate ash, high hemicellulose, and balanced lignin—which enhances hydrolysis and depolymerization under subcritical water conditions, as detailed in the decomposition pathways for lignocellulosic biomass.

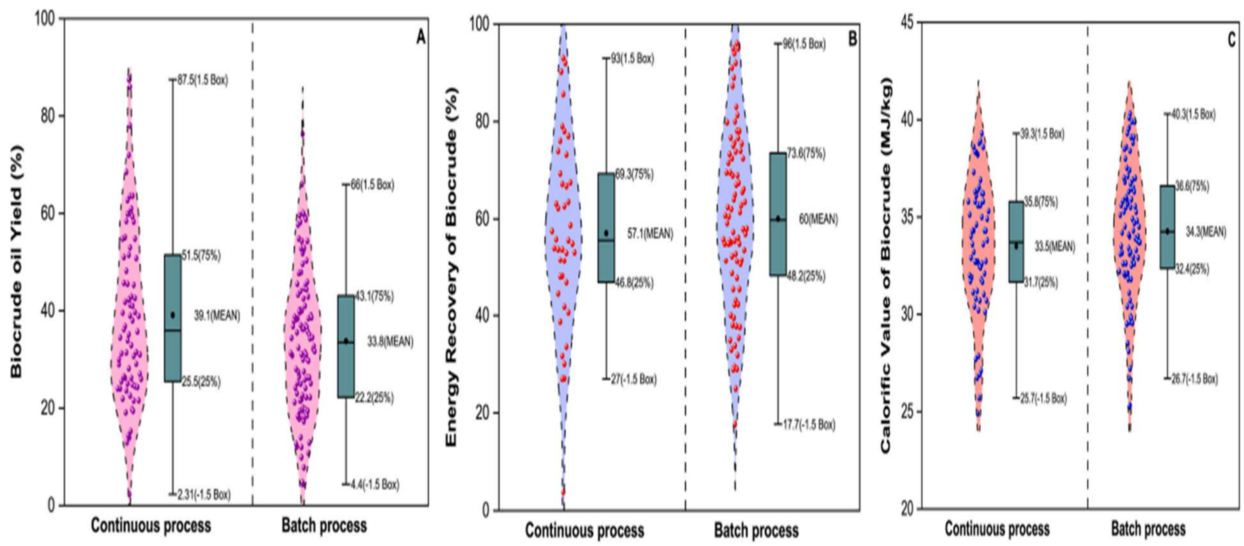

Fig 4. Kernel density diagram for (A) biocrude oil yield, (B) energy recovery, and (C) calorific value of the continuous and batch biomass HTL systems

3.2 Feedstock Composition and Bio-Crude Quality Interrelation

The ultimate analysis of the produced bio-crudes revealed a direct link between feedstock composition and fuel quality, a central theme in the reviewed literature. Bio-crude from all residues showed significant deoxygenation, with O/C ratios reduced compared to raw biomass. This improvement is a key advantage of HTL over pyrolysis bio-oil, as noted in the comparison of thermochemical methods. Specifically, the HHV of the bio-crudes (30–34 MJ/kg) is consistent with the average values reported for lignocellulosic-derived bio-crude in the violin plot (Fig 5).

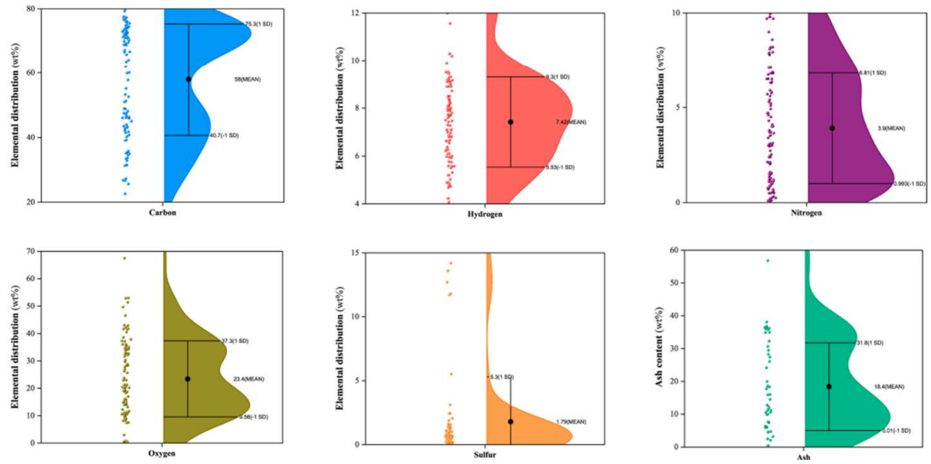

Fig 5. Violin plot showing the distribution of the physicochemical attributes of the reported biocrude oil irrespective of the HTL reaction conditions (Data obtained from Refs.

The review highlights that holocellulose (cellulose + hemicellulose) content positively influences yield, while lignin enhances the calorific value [1]. This explains the higher HHV of wheat straw biocrude, which had a significant lignin component. Conversely, the high ash content (18.7 wt%) in rice husk, primarily silica, acted as a physical barrier to conversion, leading to the lowest yield and ER. This finding underscores the critical need for feedstock selection or pre-treatment, as suggested in the review's discussion on biomass composition (Fig 6).

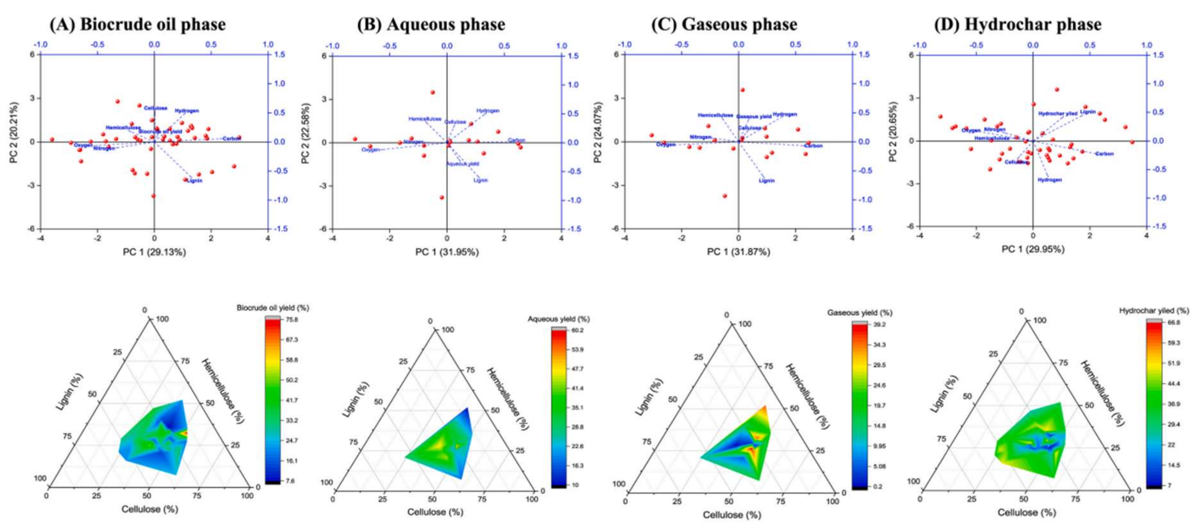

Fig 6. Principal component analysis and ternary contour diagrams showing the effect of lignocellulosic biomass composition on the product distribution and characterization (A) biocrude oil phase quantity, (B) aqueous phase quantity, (C) gaseous phase quantity, and (D) hydrochar phase quantity

The gas composition, dominated by CO_2 (55–65 vol%), is a direct result of decarboxylation reactions prevalent during HTL, as illustrated in the reaction pathways (Fig 7). The review's principal component analysis (Figs. 8 & 9) effectively visualizes how such feedstock properties dictate the final product distribution and energy output, which our experimental data substantiate.

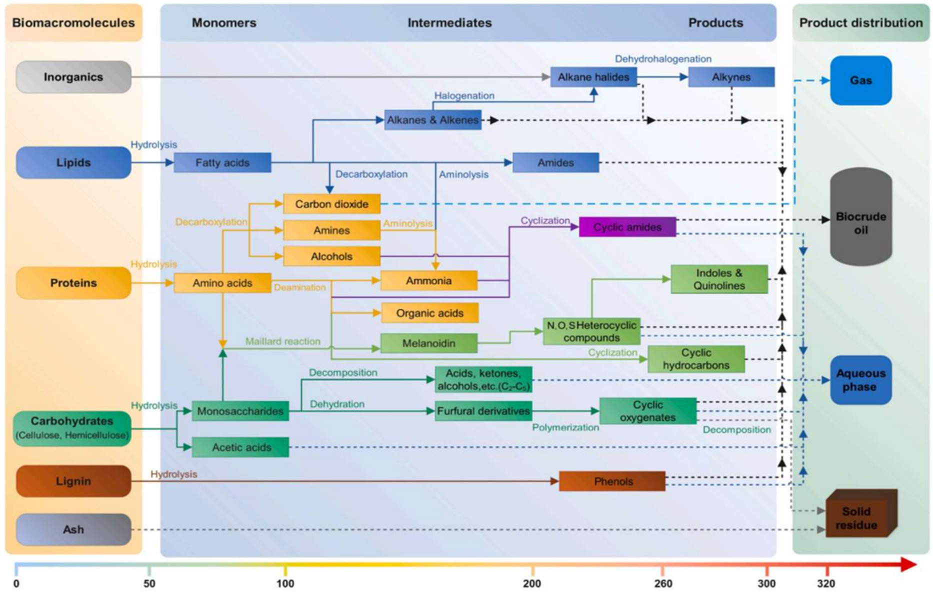

Fig 7. Potential reaction pathway of the biomass macromolecules to various end-products via HTL process as a function of temperature

3.3 Life Cycle GHG Performance and Integration into the Carbon Cycle

The Life Cycle Assessment (LCA) results demonstrate the profound emission reduction potential of the agricultural residue HTL pathway. The net negative GHG emissions (-32.1 to -47.4 g CO_2-eq/MJ) achieved through system expansion credits are a quantitative validation of the conceptual carbon cycle for modern bioenergy systems depicted in Fig. 2. This figure illustrates how bioenergy phases out fossil-derived fuels (red "X") and offers a green solution. Our LCA results translate this concept into a measurable outcome: the displacement of fossil crude and the avoidance of open-field burning create a net carbon sink. This performance is competitive with, and often superior to, the life cycle GHG emissions of biofuels from HTL reported in the literature. As shown in the comparative analysis (Fig. 8),

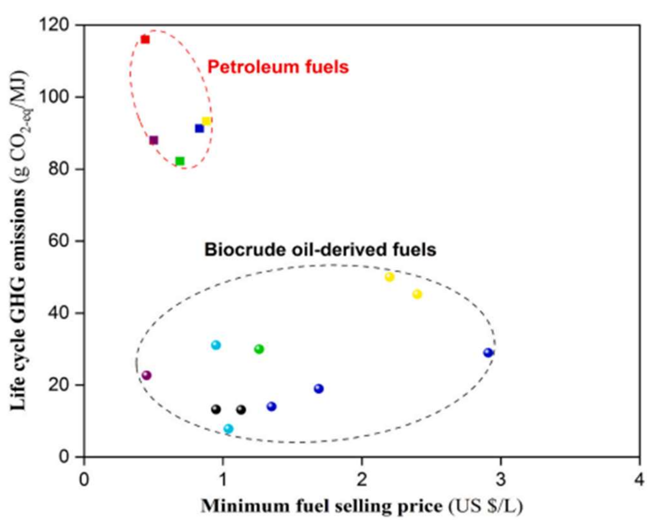

Fig 8. Comparison of minimum fuel selling prices and life cycle GHG emissions of the biofuels derived from biocrude oil with conventional petroleum fuels.

biofuels from lignocellulosic feedstocks generally exhibit lower GHG emissions than those from non-lignocellulosic feedstocks (e.g., algae). Our results for wheat straw and bagasse align with this trend, situating them favorably within the lower emission range for lignocellulosic pathways (Fig. 9).

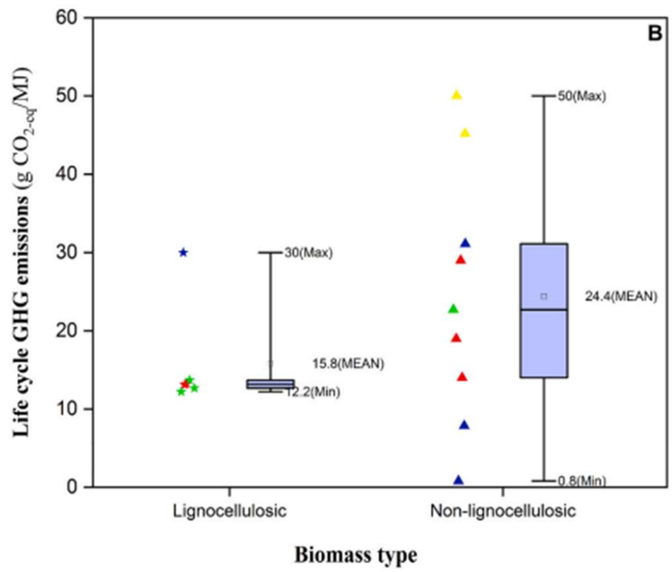

Fig 9. life cycle GHG emissions of the biofuels produced from lignocellulosic and non-lignocellulosic biomass feedstocks using the HTL process

The sensitivity analysis identifying process heat source as the most critical parameter echoes the review's emphasis on energy integration for improving techno-economic and environmental metrics [1]. Replacing grid electricity with renewable heat can reduce gross emissions by over 60%, a finding that directly supports the review's conclusion that maximizing energy efficiency and integrating renewable energy are key to commercial viability. This holistic analysis confirms that HTL of locally sourced agricultural residues is not merely a waste management strategy but a significant technological lever to address the rising global energy demand, while actively reducing net atmospheric CO_2, thereby operationalizing the sustainable carbon cycle model presented in the review.

4 CONCLUSIONS

This study conclusively demonstrates that Hydrothermal Liquefaction (HTL) of agricultural residues specifically wheat straw, sugarcane bagasse, and rice husk is a technically viable and environmentally sustainable pathway for renewable bio-crude production, with optimal conversion at 300°C yielding up to 42.5 wt% bio-crude and 78.5% energy recovery, while life cycle assessment confirms a net-negative GHG emission profile of -32.1 to -47.4 g CO_2-eq/MJ, thereby validating its potential to contribute to a circular carbon economy by displacing fossil fuels, managing agricultural waste, and supporting climate mitigation objectives.

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