Renewable and Sustainable Energy Reviews 224 (2025) 116086

Contents lists available at ScienceDirect

Renewable and Sustainable Energy Reviews

journal homepage: www.elsevier.com/locate/rser

Hydrothermal liquefaction of sewage sludge: A comprehensive review of biocrude oil production, byproducts valorization, and future perspectives

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ARTICLE INFO

Keywords:
Hydrothermal liquefaction
Strategies
Transformation pathways
Biocrude oil upgrading
Element components
Byproduct applications

ABSTRACT

Climate change is driving global efforts toward carbon neutrality and expanding renewable energy sources. Hydrothermal liquefaction (HTL) of sewage sludge offers a promising pathway for sustainable biocrude oil production. This review systematically analyzes 956 records from Web of Science and Scopus databases, with 179 articles selected for detailed analysis following PRISMA guidelines. It presents the first comprehensive and systematic analysis of biocrude oil production and byproducts valorization from the HTL of sewage sludge. Key findings highlight that mixed sludge, with a balanced organic matter composition, is ideal for biocrude oil production, achieving an average yield of 38.95 % (range: 35.3–42.6 %). Higher biocrude oil yields are more likely to be achieved under reaction conditions of approximately 350 °C and a holding time of 30 min, as indicated by 2D kernel density estimation of the collected literature. These optimal conditions are summarized as a reference point for future studies, although the exact operating conditions may need specific exploration depending on the sludge properties. The transformation of organic matter follows the order: lipids > proteins > carbohydrates > lignin/humic substances, with diverse complex reactions driving biocrude oil formation. The biocrude oil contains significant heteroatom content-nitrogen (5.5 %, range: 0.23–9.3 %), sulfur (0.9 %, range: 0–4.3 %), and oxygen (15.7 %, range: 6.7–62.8 %)-which necessitate upgrading for biocrude oil applications. Nitrogen primarily distributes into the aqueous phase, while phosphorus and metals accumulate in the solid phases, offering opportunities for resource recovery. HTL also generates byproducts in aqueous (36.67 %, range: 0.19–60.3 %), solid (22.03 %, range: 0.43–50.73 %), and gaseous (13.71 %, range: 0.2–64.68 %) phases, which can be effectively valorized through proper management, promoting both industrial applications of HTL and the development of a circular economy. This work serves as a valuable resource for researchers, policymakers, and industry stakeholders, providing insights into biocrude oil production and byproduct utilization, advancing sustainable sludge management toward global carbon neutrality goals.

1. Introduction

Addressing global climate change has spurred efforts toward achieving carbon neutrality and advancing renewable energy development. Sewage sludge, a byproduct of wastewater treatment plants, is gaining recognition as a renewable energy source, owing to its high energy content, which can reach up to 3.54 kWh/kg dry sludge [1]. In 2022, global sewage sludge production ranged from 75 to 100 million tons and is projected to rise to 130 million tons by 2030, driven by rapid population growth and industrialization [2]. However, traditional disposal methods, such as landfilling or land application, are increasingly unsustainable due to the presence of various pollutants (i.e., heavy metals, microplastics, emerging organic micropollutants), which not only waste valuable resources but also pose significant environmental and health risks [3–7]. These challenges highlight the need for innovative sludge management strategies that align with circular economy principles by transforming waste into valuable resources while minimizing environmental impacts.

Abbreviations: HTL, Hydrothermal liquefaction; COD, Chemical oxygen demand; HHV, Higher heating value; PRISMA, Preferred Reporting Items for Systematic Reviews and Meta Analysis; ATP, Attapulgite; AC, Activated carbon; HMF, 5-hydroxymethyfurfural; N/C, Nitrogen/Carbon; O/C, Oxygen/Carbon; P, Phosphorus; OP, Organic P; IP, Inorganic P; NAIP, Non-apatite inorganic P; AP, Apatite P; PAHs, Polycyclic aromatic hydrocarbons.

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https://doi.org/10.1016/j.rser.2025.116086
Received 20 December 2024; Received in revised form 23 June 2025; Accepted 9 July 2025
Available online 14 July 2025

Hydrothermal conversion technology offers a promising approach for converting biomass feedstocks into bioenergy in their wet state, avoiding the energy-intensive drying processes typically required for combustion, pyrolysis, and gasification methods [8,9]. Depending on the reaction temperature, hydrothermal conversion can be categorized into liquefaction, carbonization, and gasification, each yielding distinct end products: biocrude oil, biochar, and syngas, respectively [10]. Hydrothermal liquefaction (HTL) is particularly suited for sewage sludge due to its high moisture content and organic matter composition, enabling efficient conversion without extensive preprocessing [11]. Moreover, HTL offers dual benefits by producing renewable biocrude oil that can be further upgraded and remarkably reducing sewage sludge volume, while its byproducts (solid, aqueous, and gaseous phases (Fig. S1 and Text S1) provide opportunities for valorization, thereby enhancing the sustainability of this technology [12]. Compared to other feedstocks such as microalgae and agricultural residues, sewage sludge is more viable due to its higher energy density, abundant availability, and ease of collection [1,2]. These attributes make HTL a promising strategy for sustainable resource utilization and sludge management.

Numerous studies have been conducted to enhance biocrude oil production from HTL, examining key aspects such as feedstock selection, optimization of operation parameters, catalysts utilization, and sludge pretreatment [13–18]. However, most existing reviews have primarily concentrated on feedstocks like agricultural residues and algae [19–21], with limited attention given to sewage sludge. While some reviews do address sewage sludge, they often focus on hydrothermal carbonization, gasification, and pyrolysis, rather than HTL [22–24]. Additionally, although diverse organic matters, including lipids, proteins, carbohydrates, nucleic acids, lignin, and humic substances, play a crucial role in biocrude oil production [25–28], a systematic overview of their conversion pathways and trends in sewage sludge to biocrude oil remains lacking. Furthermore, understanding the fate and distribution of elemental components (i.e., nitrogen, phosphorus, and metals) is vital for effective biocrude upgrading and byproduct valorization [7,29,30], yet this aspect is rarely addressed in existing reviews. The management of byproducts also plays a crucial role in the industrial application of this technology [31], yet there is a lack of comprehensive summary on effective byproducts management, which is critical for maximizing resource recovery. This review aims to provide that much-needed analysis, highlighting its novelty in addressing these critical knowledge gaps.

This review presents the first comprehensive and systematic analysis of HTL of sewage sludge, conducted following PRISMA guidelines. Key strategies for enhancing biocrude oil production and the potential transformation trends and pathways of organic matter into biocrude oil are critically summarized, alongside an in-depth discussion of biocrude oil upgrading techniques. Moreover, the fate and distribution of element components (i.e., nitrogen, phosphorus, and metals) during the HTL process are thoroughly elucidated, providing insights into resource recovery opportunities. Furthermore, the potential applications of byproducts, including solid, gaseous, and aqueous phases, are explored in detail. Finally, knowledge gaps and future perspectives are highlighted. This review provides valuable guidance for advancing HTL technology, supporting industrial development, and contributing to carbon neutrality goals.

2. Material and methods

2.1. Systematic literature review on the hydrothermal liquefaction of sewage sludge

Following the guidelines of the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) [32,33], a systematic search was performed in March 2024 to identify peer-reviewed research on the HTL of sewage sludge. The search was performed using the Web of Science and Scopus databases, applying the search terms TITLE-ABS-KEY (hydrothermal liquefaction) AND TITLE-ABS-KEY (sewage sludge). The scope was restricted to conferences and research articles published between 2013 and 2024, with the publication language limited to English.

2.2. Document screening and selection and data collection

Systematic literature reviews on the HTL of sewage sludge retrieved 560 and 296 peer-reviewed articles from the Web of Science and Scopus databases, respectively (Fig. S2). After removing duplicates, the total number of records was reduced to 616 (Fig. S2). Regarding the target topic of HTL of sewage sludge, 408 documents were excluded based on title, abstract, and keyword screening for relevance, and 1 additional paper was excluded due to unavailability of the full text (Fig. S2). The remaining 207 documents underwent full-text review, resulting in the inclusion of 179 articles in this analysis. For biocrude oil data not directly available in published papers, the WebPlotDigitizer tool (https://automeris.io/wpd/) was employed to extract numerical values from graphs.

2.3. Calculations of the higher heating value of biocrude oil produced from hydrothermal liquefaction of sewage sludge

Higher heating value (HHV) is the total energy released as heat when a substance is fully combusted in oxygen, including the energy from the products measured at standard temperature of 25 °C [34]. This metric is crucial for evaluating the energy content and efficiency of fuels, including sewage sludge and biocrude oil, for various energy applications. The higher heating value was assessed using the Dulong equation [35], as illustrated in Eq. (1):

$$HHV = 0.3516 \times C + 1.16225 \times H - 0.1109 \times O + 0.10465 \times S + 0.0628 \times N \quad (1)$$

where HHV denotes the higher heating value (MJ/kg), and C, H, O, N and S, represent the mass percentage (%) of carbon, hydrogen, oxygen, nitrogen and sulfur in sewage sludge or biocrude oil discussed in this review, respectively.

2.4. Statistical analysis and data visualization

Statistical differences were evaluated using an unpaired t-test and with significance defined as $p < 0.05$ using GraphPad software. Principal Component Analysis and Partial Least Square Regression were employed to analyze the relative contributions of various parameters to biocrude oil production. Data visualization was conducted using Origin 2021, Microsoft Visio 2021, Microsoft PowerPoint Presentation, R 4.3.2, and ChemDraw 20.0.

3. Results and discussion

3.1. Strategies for enhancing biocrude oil production from hydrothermal liquefaction of sewage sludge

Previous studies have identified several key factors that significantly influence biocrude oil production from the HTL of sewage sludge. These factors include feedstock types (i.e., primary sludge, secondary sludge, etc.), operational parameters (i.e., reaction temperature, holding time, reaction medium, extraction solvents, etc.), catalysts selection and sludge pretreatment methods [13–18]. Below is a detailed summary of these factors.

3.1.1. Selection of feedstock types

Among the 179 analyzed articles, the distribution of studies across different feedstocks was as follows: sewage sludge (61), secondary sludge (14), digested sludge (12), mixed sludge (8), and primary sludge

(7) (Fig. 1A). Biocrude oil yields followed the order: mixed sludge > primary sludge > secondary sludge > corresponding digested sludge. The yields for mixed sludge and primary sludge were significantly higher than those for secondary sludge (p = 0.0070–0.0226), while mixed sludge showed a slightly higher yield compared to primary sludge (p = 0.5114). Specifically, the yields ranged from 35.3 % to 42.6 % for mixed sludge, 29.8 %–46.24 % for primary sludge, and 22.11 %–30.7 % for secondary sludge, with respective averages of 38.95 ± 5.16 %, 35.03 ± 6.95 %, and 25.39 ± 3.20 % (Fig. 2A). Notably, the positive synergistic effects of mixed sludge, resulting from its balanced organic composition, can be attributed to Maillard reactions, where the higher protein content in secondary sludge provides amino groups that interact with oxygen-containing molecules from primary sludge, thereby enhancing biocrude oil yields [38]. Primary sludge generally outperforms secondary sludge in biocrude oil production, likely due to the higher ash content in secondary sludge, which can inhibit organic matter conversion during HTL [13,39]. Additionally, non-digested sludge yielded significantly more biocrude oil than digested sludge (p = 0.0006), with average values of 29.79 ± 5.44 % (range: 22.79–40.83 %) and 18.43 ± 5.92 % (range: 9.64–27.64 %), respectively (Fig. 2A). This difference is likely due to the more recalcitrant nature of digested sludge, which makes it less amenable to degradation during HTL [40].

In contrast, some studies (20 %) have reported opposing trends. For instance, Kulikova et al., reported that secondary sludge produced a higher biocrude oil yield (30.7 %) compared to primary sludge (29.8 %). This discrepancy may be attributed to differences in sludge composition, particularly the higher ash content (33.2 % versus 24.9 %) and lower lipid content (5.2 % versus 10.2 %) in secondary sludge compared to primary sludge in this study. Moreover, Barreiro et al., found that non-digested sludge and digested sludge exhibited similar biocrude oil yields (27.36 % versus 27.64 %) under HTL conditions of 320 °C 10 min (11.1 %). The main reason for this phenomenon is likely due to the relatively small variation in the physiochemical properties of the sludge, including water content and surface characteristics, despite some degree of organic matter degradation.

3.1.2. Optimization of operating parameters

Optimizing operational parameters is the most commonly employed strategy for enhancing biocrude oil production from the HTL of sewage sludge [15,42–44] as evidenced by 69 studies, far surpassing investigations into catalysts selection (n = 26) and pretreatment methods (n = 7) (Fig. 1A). Among these studies, considerable attention has been devoted to key operational parameters, including temperature (n = 57), holding time (n = 33), reaction medium (n = 11), and extraction solvents (n = 7) (Fig. 1B). These findings highlight the emphasis on optimizing process conditions to maximize biocrude oil yields, as detailed below.

3.1.2.1. Temperature. The relationship between the reaction temperature and biocrude oil yields in HTL is illustrated in the kernel density plot (Fig. 3A). The density values peak at 350 °C (2.37E-04), which was significantly higher than lower (e.g., 200 °C, density = 1.04 E−9) and higher (e.g. 400 °C, density = 2.83E-06). Compared to the lower temperature, the enhanced biocrude oil production observed at elevated temperatures (below 350 °C) can be attributed to two key factors: 1) accelerated hydrolytic reaction and improved solubility of hydrophobic organic matter due to decreased density, polarity, and dielectric of water [45], and 2) the conversion of fatty acids into alkanes or alkenes through decarboxylation or their transformation into biocrude oil constituents via cross-linking reactions, such as amide formation with proteins [46]. However, the decline in biocrude oil yields observed at reaction temperatures above 350 °C can be attributed to the conversion of biocrude oil components into gaseous or aqueous products through cracking reactions, as well as the formation of solid residues with high molecular weight through repolymerization [47,48]. Notably, Wang et al., reported that the maximum biocrude oil production was achieved at 270 °C, representing a 16.24 % increase compared to the yield at 320 °C (40.82 %), which contrasts with the findings of this study. This discrepancy can primarily be attributed to reduced temperature requirements facilitated by stirring [49], along with the use of catalysts and the variations in reduction media during HTL [47].

3.1.2.2. Holding time. The impact of holding time on biocrude oil yields from HTL of sewage sludge is presented in Fig. 3B. The results showed that as holding time increased, biocrude oil yields initially exhibited an upward trend, follow by a gradual decline. Specifically, the yield density increased from 7.62E-9 to a peak of 7.91E-4 at approximately 30 min of holding time, after which it steadily decreased to 2.41E-14 (Fig. 3B). These findings suggest that setting the holding time for HTL to approximately 30 min offers remarkable potential for achieving higher

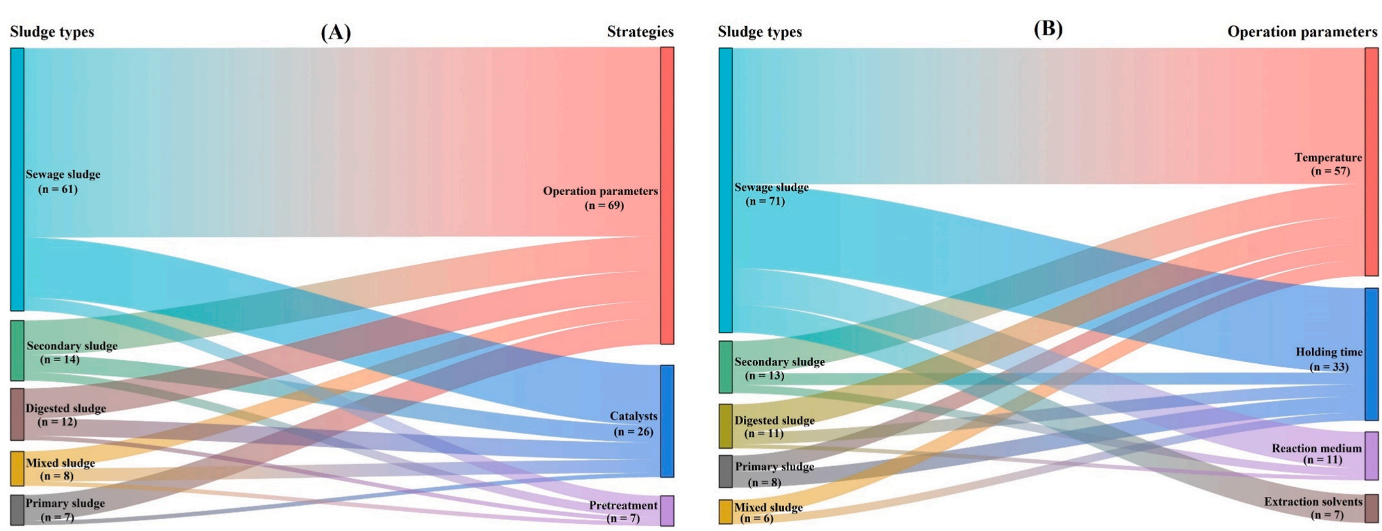

Fig. 1. Sankey plot of sludge types and optimization strategies in hydrothermal liquefaction (A) and key operational parameters for biocrude oil production (B). Note: The number of sludge types differs between (A) and (B) because some studies address multiple operational parameters within the same research. The digit (n = *) illustrates the number of research articles focused on each strategy/parameter. Additionally, "sewage sludge" refers to studies where the specific sludge type is not explicitly mentioned, with the feedstock broadly described as sewage sludge or municipal sludge. Notably, only categories with more than five studies are included in the figure to ensure clarity and avoid overinterpretation or insignificance from limited data.

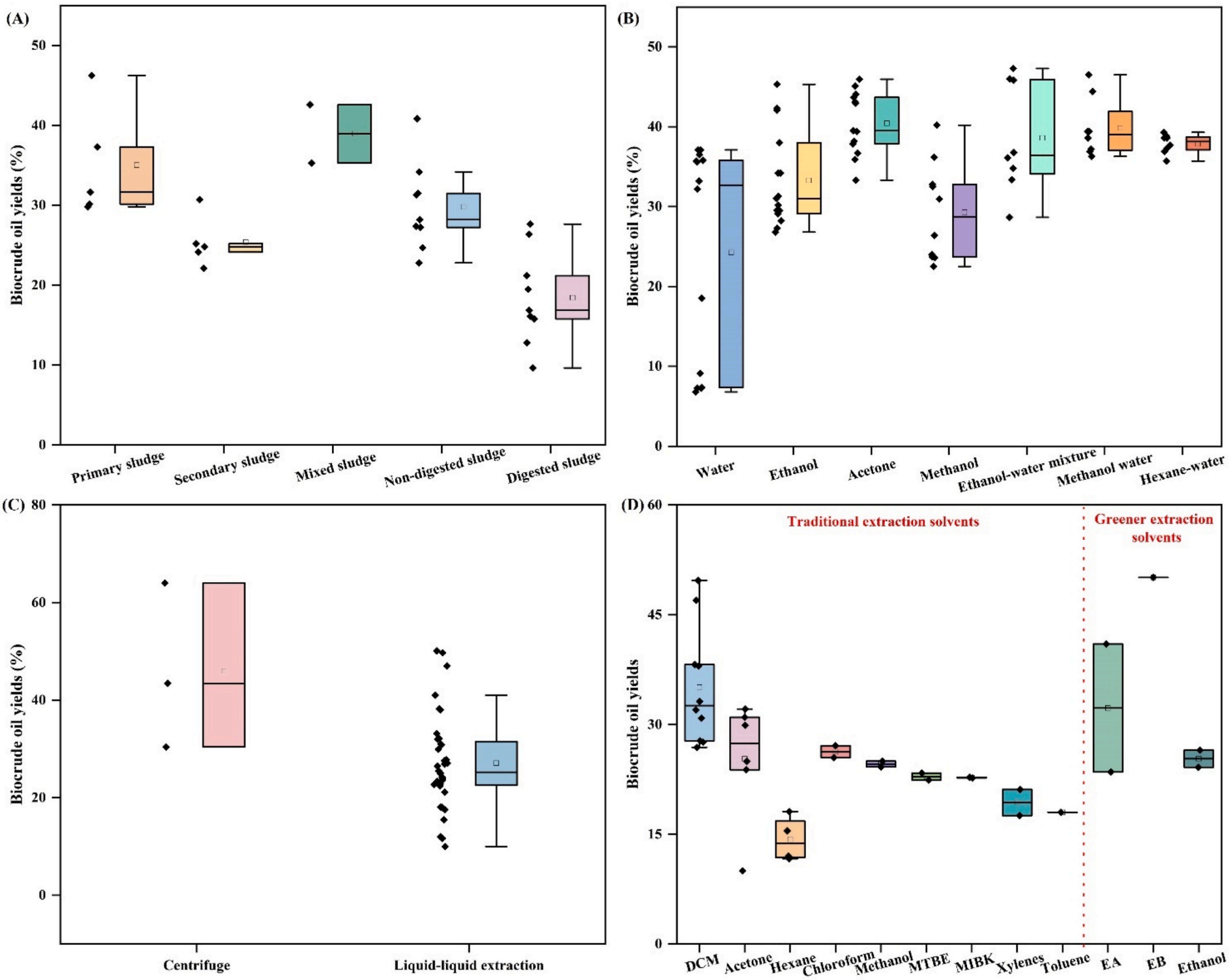

Fig. 2. Box plot of biocrude oil yields in hydrothermal liquefaction from various sludge types (A), reaction mediums (B), extraction methods (C), and solvents used for liquid-liquid extraction (D). Note: Mixed sludge refers to a combination of primary and secondary sludge, while the difference between non-digested sludge and digested sludge lies in whether the sludge undergoes the anaerobic digestion process prior to hydrothermal liquefaction. For the original data used to create these figures, please refer to Table S1, Table S5 and Table S6 in the Supplementary Materials. DCM, MTBE, MIBK, EA, and EB represent dichloromethane, methyl isobutyl ketone, methyl tert-butyl ether, ethyl acetate, and ethyl butyrate, respectively.

biocrude oil yields. At the onset of the HTL process (from 0–30 min), sewage sludge is initially broken down into smaller components, which explains why extending the holding time properly contributed to the increased yield of biocrude oil [50]. However, after a certain duration (i. e., over 30 min), the likelihood of repolymerization, cyclization, and condensation reactions among intermediate products increases, leading to higher hydrochars and gaseous products formation, which in turn reduces the biocrude oil yield [51]. In contrast, Obeid et al., reported that the holding time of 60 min was more effective than 30 min in promoting biocrude oil accumulation. This difference can be attributed to the HTL experiments being conducted at 300 °C, where recalcitrant organic matter could not be effectively degraded. Extending the holding time facilitated the breakdown of organic matter, thereby enhancing biocrude oil production.

To elucidate the relative contributions of reaction temperature and holding time to biocrude oil production during the HTL of sewage sludge, the Principal Component Analysis (Fig. 4) and Partial Least Square Regression (Fig. S3) were performed. The Principal Component Analysis results revealed that reaction temperature and holding time accounted for 54.35 % and 45.65 % of the variance in biocrude oil yields, respectively (Fig. 4). This suggests that optimizing reaction temperature has a greater impact on enhancing biocrude oil yields compared to holding time. The major contribution of the reaction temperature is also supported by Partial Least Square Regression (Fig. S3), with detailed analysis provided in Text S2. These findings are consistent with previous studies [16,36].

3.1.2.3. Reaction medium. A total of 11 studies investigated the impact of reaction medium on biocrude oil yields (Fig. 1B), with 89 data points analyzed. The results are summarized in the box plot (Fig. 2B). Commonly used reaction mediums for HTL include water (18 data points), organic solvents (47 data points; ethanol = 24, acetone = 13, methanol = 10), and cosolvents (25 data points; ethanol-water = 8), methanol-water = 8), and hexane-water = 8) (Fig. 2B). Except for methanol, all organic solvents and cosolvents remarkably enhanced (p = 0.0004–0.0276) biocrude oil yields compared to pure water. Among the organic solvents analyzed, acetone exhibited the highest biocrude oil yields (range: 33.3–45.95 %; average 40.44 ± 3.96 %), followed by ethanol (range: 26.8–45.31 %; average 33.27 ± 5.95 %), and methanol (range: 22.5–40.2 %; average 29.28 ± 6.13 %), with yields 1.21–1.67

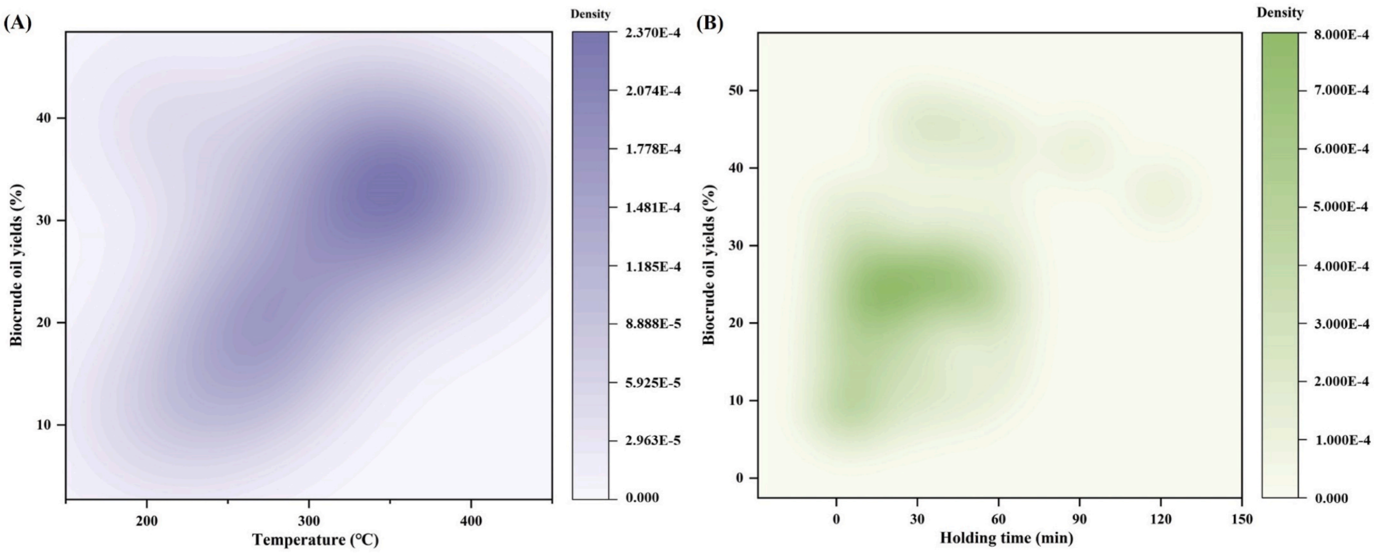

Fig. 3. 2D Kernel density plots of biocrude oil yields under varying hydrothermal liquefaction conditions: temperatures (A) and holding times (B). Note: Higher density values correspond to greater biocrude oil yields. A total of 117 data points were included in the analysis. For the original data used to create these two figures, please refer to Table S2-S3 in the Supplementary Materials.

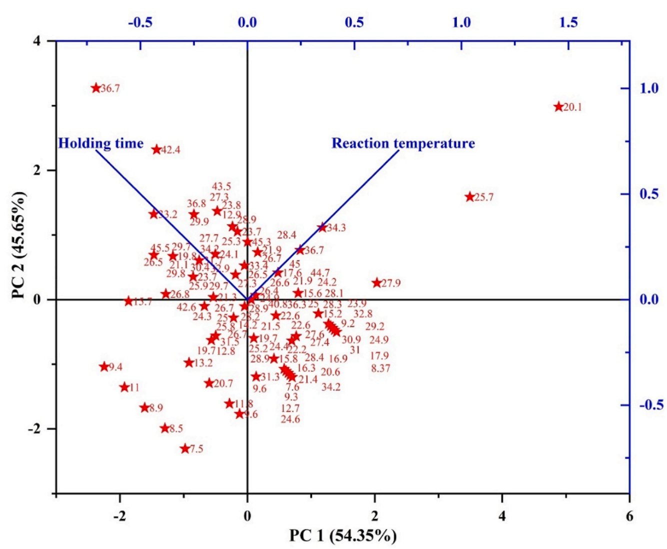

Fig. 4. The relative contribution analysis of temperature and holding time on biocrude oil production during the hydrothermal liquefaction of sewage sludge through Principal Component Analysis. The data supporting this analysis is provided in Table S4 of the Supplementary Materials.

times higher than water (range: 6.79–37.1 %; average 24.25 ± 13.72 %) (Fig. 2B). The superior performance of organic solvents can be attributed to their role as reaction substrates, interacting with decomposition intermediates to promote biocrude oil formation [52,53]. Moreover, the higher molecular weights of acetone (58) and ethanol (46) compared to methanol (32) likely explain their greater effectiveness as reaction mediums [54]. These findings align with previous studies indicating that acetone and ethanol are more effective than methanol in enhancing biocrude oil yields [44,52]. Despite the higher yield resulting from acetone, ethanol emerges as a more promising solvent because: 1) biocrude oil from acetone contains primarily N-containing compounds and ketones, resulting in a lower calorific value (26.74 MJ/kg), compared with the ester compounds (similar to biodiesel, 38.42 MJ/kg) derived from ethanol [52]; 2) ethanol can be sourced from biomass bioconversion, making it a greener option compared to acetone [55]; and 3) ethanol has lower toxicity and is safer to handle compared to acetone, reducing environmental and health risks during processing [44].

Additionally, among the cosolvents analyzed, these mixed solvents demonstrated comparable performance in promoting biocrude oil yields ($p = 0.1706$–0.7694), with yields remarkably higher than those obtained using water alone as the reaction mediums (p = 0.0054–0.0124). Specifically, biocrude oil yields ranged from 36.3 to 46.5 %, 28.65–47.29 %, and 35.7–39.3 % for methanol-water, ethanol-water, and hexane-water mixtures, respectively, with average yields of 39.84 ± 3.69 %, 38.6 ± 6.9 %, and 37.86 ± 1.19 (Fig. 2B). The superior performance of organic solvent-water mixtures compared to using either organic solvents or water alone can be attributed to synergistic effects. These effects arise from the enhanced solubility of reactants, intermediates, and products in the mixed medium, facilitating mass transfer and promoting reaction kinetics [56,57]. Meanwhile, the co-solvent system may balance the polarity of the reaction medium, optimizing the breakdown of complex organic molecules and improving biocrude oil formation efficiency [58]. Notably, the findings of this study all align closely with previous studies on the effectiveness of co-solvents in enhancing biocrude oil yields, further reinforcing the validity of these synergistic mechanisms.

3.1.2.4. Extraction methods. The extraction process plays a pivotal role in determining biocrude oil yields, as highlighted by seven studies comprising 39 data points (Figs. 1B and 2C). Extraction methods can be broadly categorized into two types: centrifuge-based methods and liquid-liquid extraction (Fig. 2C). Statistical analysis shows that centrifuge-based methods significantly outperform liquid-liquid extraction (p = 0.0005), yielding 1.70 times higher biocrude oil yields. Specifically, biocrude oil yields ranged from 30.38 % to 64 % with centrifuge-based methods and from 10 % to 50.1 % with liquid-liquid extraction, with average yields of 45.94 ± 16.95 % and 27.08 ± 9.72 %, respectively (Fig. 2C). The superior biocrude oil recovery of centrifuge-based methods stems from their ability to create a distinct oil-water interface for efficient phase separation while effectively extracting residual oil with minimal solvent consumption [15,43]. The findings from both collected studies align with this observation. However, it is noteworthy that centrifuge-based methods are generally associated with high energy consumption, which could limit their industrial application. Hence, the recovery efficiency of solvents used in liquid-liquid extraction was further analyzed, with the results presented in Fig. 2D.

Liquid-liquid extraction solvents in reported studies can be categorized as traditional (i.e., dichloromethane, acetone, hexane, etc.) or greener (i.e., ethyl acetate, ethyl butyrate, etc.) (Fig. 2D). Among traditional solvents, dichloromethane exhibited superior performance, yielding 1.39–1.95 times more biocrude oil than other solvents (p = 0.0002–0.025; significance analysis not conducted for fewer than two data points). This can be attributed to its broad extraction range, including fatty acids, non-cyclic and cyclic oxygenates, ester derivatives, etc. [59]. For greener extraction, ethyl butyrate showed the highest efficiency (biocrude oil yield up to 50.1 %), 1.55–2.78 times higher than others (Fig. 2D), likely due to its nonpolar nature, aligning with the predominantly nonpolar composition of sewage sludge [60,61]. Notably, greener solvents outperformed traditional ones, not only in extraction efficiency but also in producing biocrude oil with lower heteroatom content (i.e., N and S), reducing the need for extensive upgrading [61].

3.1.3. Utilization of catalysts during hydrothermal liquefaction

A total of 26 articles investigated the use of catalysts for biocrude oil yield from the HTL of sewage sludge (Fig. 1A), yielding 106 data points (Fig. 5). These catalysts can be classified into acidic and basic catalysts based on their acid-base characteristics, as well as homogeneous and heterogeneous catalysts based on their phase relationship with the reactants. Among these, heterogeneous catalysts attracted the most attention, with 66 data points, followed by basic (22), homogeneous (12), and acidic catalysts (6) (Fig. 5). The main reason for this preference lies in the superior characteristics of heterogeneous catalysts, which include excellent stability during HTL, high selectivity that enables efficient control over reaction pathways, reduced byproduct formation, and lower operational costs due to their ease of recovery through simple methods (i.e., filtration or centrifugation).

All catalyst types improved biocrude oil yield, with acidic catalysts outperforming basic catalysts and heterogeneous catalysts more effectively than homogeneous ones (Fig. 5). The average yields were 27.47 ± 7.83 % (p = 0.2029) for acidic catalysts and 28.73 ± 8.81 % (p = 0.3354) for basic catalysts, showing 1.28- and 1.10-fold improvements, respectively, compared to control groups (Fig. 5A). Acidic catalysts likely enhanced reactant activity, promoting organic matter degradation [37]. However, one data point (16.67 %) contradicted this, where acidic catalyst (i.e., Al_2O_3) seemed to favor undesirable reactions, leading to more solid residue formation and a lower biocrude yield [62]. Additionally, basic catalysts likely raised pH, enabling the conversion of carbohydrates into intermediate components via decomposition, depolymerization, dehydration, and decarboxylation into long-chain hydrocarbons [63–65]. Thus, basic catalysts are recommended for sludge with high carbohydrate content, as higher carbohydrate levels may more effectively activate the catalyst, increasing biocrude yield.

The promotion effects of heterogeneous catalysts were slightly greater than those of the homogeneous catalysts (Fig. 5B). The average yields achieved were 30.54 ± 15.55 % (p = 0.3828) for heterogeneous catalysts and 28.91 ± 11.18 % (p = 0.7771) for homogeneous catalysts, corresponding to 1.08- and 1.04-fold increases compared to the respective control groups (Fig. 5B). The enhancement is primarily attributed to the activity of cations (i.e., Cu, Fe, Co), which facilitate key reactions during HTL [42]. However, 26 data points (39.4 %) for heterogeneous catalysts and 4 data points (33.3 %) for homogeneous catalysts exhibited a negative impact on biocrude oil yield during HTL. This discrepancy may stem from variations in sludge characteristics, reaction solvents, or extraction methods, which could influence the overall process efficiency and outcomes [17,66].

The impact of support materials and the use of single or bimetallic catalysts, essential components of heterogeneous and homogeneous systems, is further analyzed, with results presented in Fig. 5C and D. ATP (attapulgite) and HZSM-5 exhibited superior performance in facilitating biocrude oil yields, achieving 1.45–1.47 fold increases, whereas Al_2O_3 and AC (activated carbon) showed modest improvements of 1.06–1.19-fold compared to controls (Fig. 5C). These results are attributed to their superior properties, including high surface area, porosity, strong acidity (in the case of HZSM-5), excellent thermal stability, and efficient metal-support interactions, which collectively enhance reaction efficiency and selectivity while reducing byproduct formation [62,67,68]. Moreover, biocrude oil yields were slightly higher with bimetallic catalysts compared to single-metal catalysts (29.96–37.28 % versus 23.65–28.78 %, Fig. 5D), primarily due to the ability of bimetallic catalysts to provide more active sites for hydrogenation reactions, enhancing catalytic efficiency and product yields [62,69].

3.1.4. Implementation of sludge pretreatment methods prior to hydrothermal liquefaction

Seven studies explored sludge pretreatment methods prior to HTL to enhance biocrude oil yields (Fig. 1A), yielding a total of 38 data points, with the outcomes presented in Fig. 5E. The results indicated that sludge pretreatment, encompassing physical, chemical, and combined approaches, significantly enhanced biocrude oil yields (p = 0.0001–0.049). Among these, combined pretreatment demonstrated the highest efficacy, followed by chemical pretreatment, while physical treatment exhibited the least improvement (Fig. 5E). The average biocrude oil yields through combined, chemical, and physical pretreatment methods were 35.66 ± 7.86 %, 28.27 ± 6.42 %, and 27.35 ± 4.44 %, respectively, representing 1.54-, 1.31-, and 1.16-fold increases compared to the control groups (Fig. 5E). These improvements stem from the disruption of microbial cells and sludge floc structure, enhancing the availability of cellular organic matters for hydrothermal processing [18,27,70]. Generally, the disruption effectiveness follows the trends: combined > chemical > physical, aligning with the observed biocrude oil yield variations [18,27]. Additionally, certain pretreatments (i.e., acids) dissolve alkali metals and reduce ash content, mitigating inhibitory effects on HTL, and further boosting biocrude oil production [11]. Notably, currently employed methods (i.e., ultrasonic, microwave, NaClO addition, etc.) are often associated with high energy and chemical consumption, which increases operational costs for wastewater treatment plants and limits their feasibility for large-scale industrial applications [18,27,70].

3.2. Potential transformation of organic matters in sewage sludge for biocrude oil production via hydrothermal liquefaction

Biocrude oil from HTL of sewage sludge is produced by the thermal breakdown of organic matter under high temperature and pressure in a water-rich environment. Sewage sludge is rich in lipids, proteins, carbohydrates, lignin, nucleic acid, etc. [71], holds up to 3.54 kWh/kg of energy in dry weight, making it a promising renewable energy source [2]. This section reviews the key transformation trends and pathways of organic matter in sewage sludge during HTL and classifies the resulting biocrude oil components.

3.2.1. Potential transformation trends and pathways of organic matter in sewage sludge to biocrude oil

The transformation of organic matter during HTL for biocrude oil production follows the order: lipids, proteins, carbohydrates, and lignin/humic substances. Model compound studies reported energy recovery rates at 350 °C of 82.7–86.7 % for lipids, 10.7–36.4 % for proteins, 8.3–13.7 % for carbohydrates, and 2.5 % for lignin and/or humic substances [26,72,73]. Similarly, Teri et al., observed biocrude yields of >90 % for lipids, ~30–35 % for proteins, and 10–15 % for carbohydrates [74]. Interestingly, mixtures often matched mass-average yields, but synergistic effects were noted in polysaccharide-protein combinations under severe conditions, resulting in higher than expected yields, contributed to Millard reactions [26]. In wastewater treatment plants, primary sludge (rich in lipids) and secondary sludge (rich in proteins) are typically produced [75,76], combining these sludges creates a balanced feedstock for HTL. However, the extent of synergistic enhancement depends on both the feedstock composition and HTL

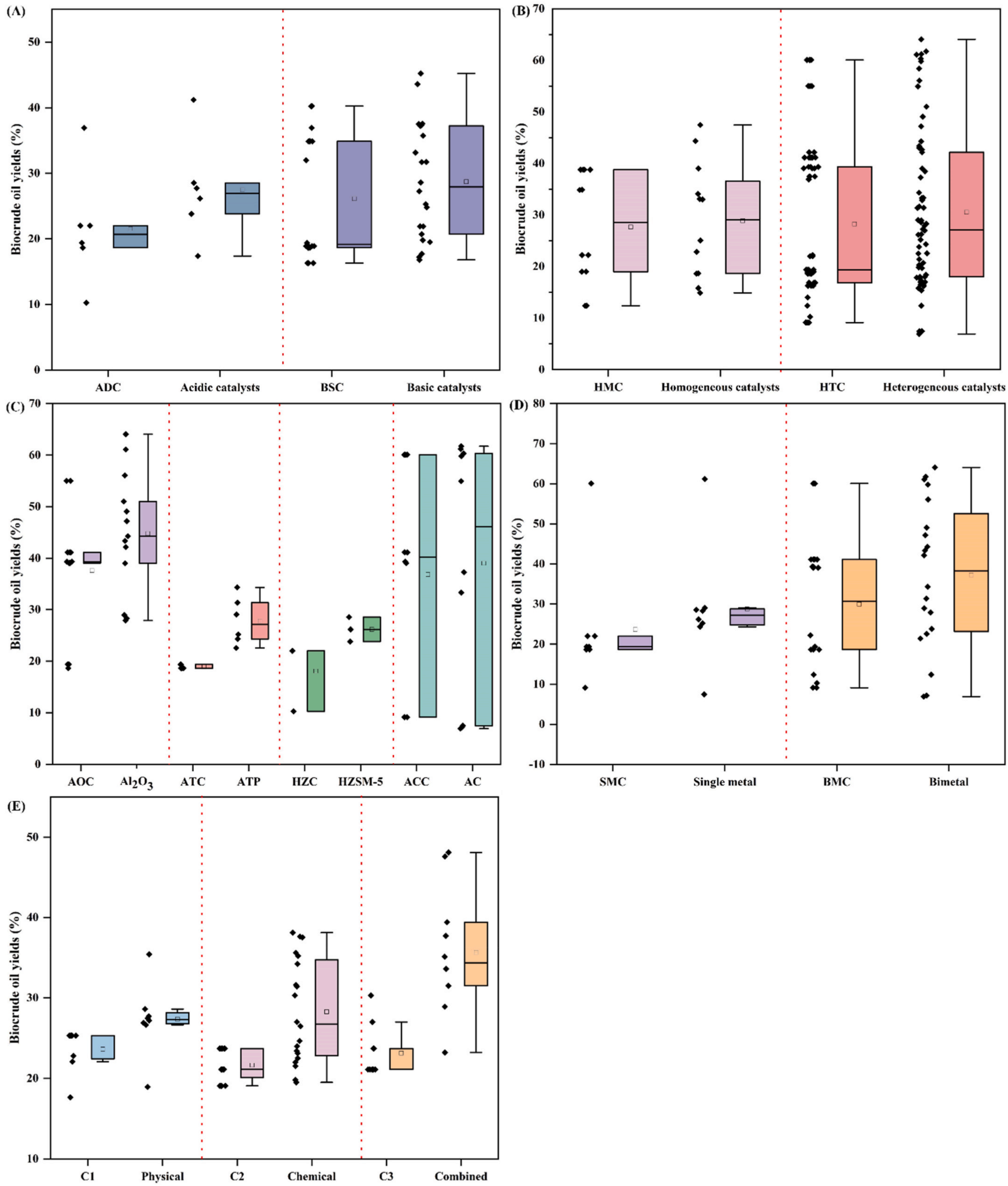

Fig. 5. Box plot of biocrude oil yields from the hydrothermal liquefaction of sewage sludge under various catalytic conditions (A–D), including the effects of acidic and basic catalysts (A) and homogeneous and heterogeneous catalysts (B), support material types (C) and single versus bimetallic catalysts (D), alongside the influence of sludge pretreatment methods prior to liquefaction (E). Note: ADC, BSC, HMC, and HTC represent the control groups for acidic, basic, homogeneous, and heterogeneous catalyst addition, respectively, in biocrude oil yield assessment.Note: AOC, ATC, HZC, ACC, SMC, NMC represent the control groups for Al_2O_3, ATP (attapulgite), HZSM-5, AC (activated carbon), single metal, and bimetallic, respectively, in biocrude oil yield assessment. C1, C2, and C3 represent the ocontrol groups for physical, chemical, and combined pretreatment methods of sludge prior to liquefaction. For the original data used to create this figure, please refer to Table S7 and Table S8 in the Supplementary Materials.

conditions [77,78], highlighting the importance of process optimization for efficient HTL application in wastewater treatment plants.

The formation pathways for converting organic matter in sludge to biocrude oil through HTL are summarized in Fig. 6. Lipids hydrolyze into glycerol and fatty acids, forming key biocrude oil precursors [27] (Fig. 6). Glycerol produces alcohols, acids, and aldehydes through decomposition, while fatty acids generate amides, alkenes, alkanes, and naphthalene through amidation, decarboxylation, and aromatization processes, collectively contributing to biocrude oil yields [25]. Notably, cholesterol and its derivates (i.e., cholestenone and cholestene) are detected in municipal sludge [29,79] but contribute minimally due to low cholesterol content [26] (Fig. 6). Proteins hydrolyze into amino acids, which undergo reactions like decarboxylation, deamination, cyclization, decomposition, lactamization, and Maillard reactions (Fig. 6) [26]. These processes generate key compounds, including amines, ketones, aldehydes, piperidine, phenolic compounds, and caprolactam, all of which contribute to biocrude oil composition [27,80].

Carbohydrates hydrolyze into monomers like glyceraldehyde, pentose, and hexoses, which then dehydrate into intermediates such as pyruvaldehyde, 5-hydroxymethylfurfural (HMF), and furfural (Fig. 6). These intermediates undergo rehydration, dehydration, decomposition, rearrangement and oligomerization, producing acids (i.e., lactic, acetic, levulinic), alcohols (i.e., cresol, phenol), and cyclopentanone, which are components of biocrude oil (Fig. 6). Maillard reactions between amino acids and carbohydrates generate nitrogen-containing heterocycles, such as pyrazine, pyrrole, indole, and pyridine, which also contribute to biocrude oil components [27,80]. Lignin undergoes hydrolysis to form phenolic compounds and alcohols, which then produce benzene, naphthalene, and polycyclic aromatic hydrocarbons (PAHs) through hydrogenation and deoxidation, dimerization, hydrogen abstraction and acetylene addition [25], all of which contribute to biocrude oil composition (Fig. 6). Notably, lignin and humic substances derived products contribute minor amounts to biocrude oil [25,81,82]. Nucleic acids hydrolyze into nucleotides, ribose, and phosphate groups, with nitrogenous bases undergoing deamination, releasing ammonia as a gas-phase product [28].

3.2.2. The classification of biocrude oil components

The detailed analysis of the potential formation pathways for converting organic matter in sludge to biocrude oil through HTL is presented in Section 3.2.1. Building on this, a comprehensive examination of the components of biocrude oil derived from HTL end products is examined. The findings indicate that the components of the biocrude oil phase can be classified into three major categories: nitrogenated compounds, oxygen-containing compounds, and hydrocarbons. Specifically, nitrogenated compounds include amines, amides, piperidine, caprolactam, and nitrogen-containing heterocyclic compounds such as pyrazine, pyrrole, indole, and pyridine. Oxygen-containing compounds encompass ketones, aldehydes, phenolic compounds (including phenol derivatives), alcohols, acids, esters, and cyclopentane. Lastly, hydrocarbons consist of benzene, naphthalene, light PAHs, heavy PAHs, alkenes, and alkanes. These classifications align with the findings of Shah et al., who similarly categorized biocrude oil components in their study.

Elemental composition (C, H, O, N, S) of biocrude oil from HTL of sewage sludge shows wide variability: C (27.6–76.9 %), H (4.4–11.6 %), N (0.23–9.3 %), S (0–4.3 %), and O (6.7–62.8 %), with averages of 68.3, 8.9, 5.5, 0.9, and 15.7 %, respectively (Fig. 7A–E). Only 17 % of the H data and 3.4 % of the N data meet the petroleum crude specifications (83–87 % C, 10–14 % H, 0.1–1 % N, and 0.1–3 % O) [47], while none of the C and O data align (Table S9). The high average contents of N and O observed in biocrude oil across all collected data points appears to be primarily associated with HTL of high-protein feedstock. However, it is important to clarify that this review focused on studies involving sewage sludge regardless of the protein content of the feedstock. In the collected literature, the number of studies involving secondary sludge as the feedstock for HTL far exceeds those using primary sludge (Fig. S5). Given that secondary sludge typically contains significantly higher protein content than primary sludge, this likely contributes to the elevated N and O levels observed in biocrude. Moreover, even when considering primary alone, the average N and O contents in the resulting biocrude oil was 4.23 % and 9.36 % (Fig. S5), respectively, both of which significantly higher than the typical specification for petroleum crude (0.1–1 % for N and 0.1–3 % for O). Therefore, this does not impact the overall conclusion that high heteroatom content in biocrude oil necessitates further upgrading to enable its potential industrial application.

Additionally, the high content of heteroatoms in biocrude results in lower energy recovery, with higher heating values ranging from 8.62 to 39.21 MJ/kg, averaging 33.07 MJ/kg, remarkably lower than the 42.8 MJ/kg of petroleum crude (Fig. 7F). Further analysis of nitrogen/carbon (N/C) and oxygen/carbon (O/C) ratios is conducted, presented in Fig. S4, reveals that none of the research data align with the conventional petroleum standards (N/C: 0.001–0.01; O/C: 0.01–0.05) [59] (Fig. S4). This is primarily due to the formation of oxygen functional groups from the decomposition of carbohydrates and proteins, as well as the nitrogen and oxygen heterocycles generated via Millard reactions [26]. High N/C and O/C ratios can result in decreased combustion performance, reduced thermal stability, increased corrosiveness, catalyst poisoning, processing difficulties and a decline in the quality of final products, ultimately impacting market competitiveness and environmental sustainability [40,84–86]. Therefore, it is essential to improve the fuel quality of biocrude oil through various strategies, which will be discussed in detail in the following sections.

3.3. Upgrading of biocrude oil derived from hydrothermal liquefaction of sewage sludge

As discussed in Section 3.2.2, biocrude oil contains a high concentration of heteroatoms, primarily oxygen and nitrogen (N_xO_y species) [87], with average contents of N (5.5 %), and O (15.7 %) (Fig. 7C and E). These values are based on statistical analysis rather than single data points and significantly exceed those found in petroleum crude oil, where nitrogen and oxygen contents typically range from 0.1 to 1 %, and 0.1–3 %, respectively [47]. Moreover, the biocrude oil contains highly unsaturated molecules with high molecular weights, such as aromatic hydrocarbons, polycyclic aromatic hydrocarbons, and heterocyclic compounds [88]. This composition can lead to undesired properties, such as high acidity, increased viscosity, poor mechanical behaviors upon aging, and a tendency for polymerization [40,89,90]. Therefore, effective removal of heteroatoms is critical to meet the requirements for downstream upgrading and utilization of biocrude oil.

Hydrotreatment is the most widely used for biocrude oil derived from sewage sludge, removing S, N, and O through hydrodesulfurization, hydrodenitrogenation, and hydrodeoxygenation, respectively [91–93]. This process is generally achieved at 200–450 °C, and improves the quality of the biocrude oil by breaking the heteroatom-carbon bond, saturating hydrocarbons, and cracking heavy molecules into lighter ones [88,94]. This eventually enhances calorific value and hydrogen-to-carbon ratio of the biocrude oil. For instance, hydrotreatment of biocrude oil derived from digested sludge achieved heteroatom removal rates of up to 99.1 % (S), 96.6 % (O), and 91 % (N) and increased HHV from 36.3 MJ/kg for raw biocrude oil to 45.9 MJ/kg [88]. However, biocrude with high nitrogen content can destabilize under severe conditions (~400 °C), making two-stage hydrotreatment necessary [95]. Previous studies reported a two-stage hydrotreating process, where initial deoxygenation was achieved at 350 °C with minimal coke formation, followed by up to 92 % nitrogen removal at 400 °C in the second stage, effectively reducing coke yield by 3.4-0.7 % compared to single-stage treatment [95]. This two-stage approach effectively upgrades high-nitrogen biocrude oil to meet fuel standards, aligning with the findings of Heracleous et al.,. In addition, although

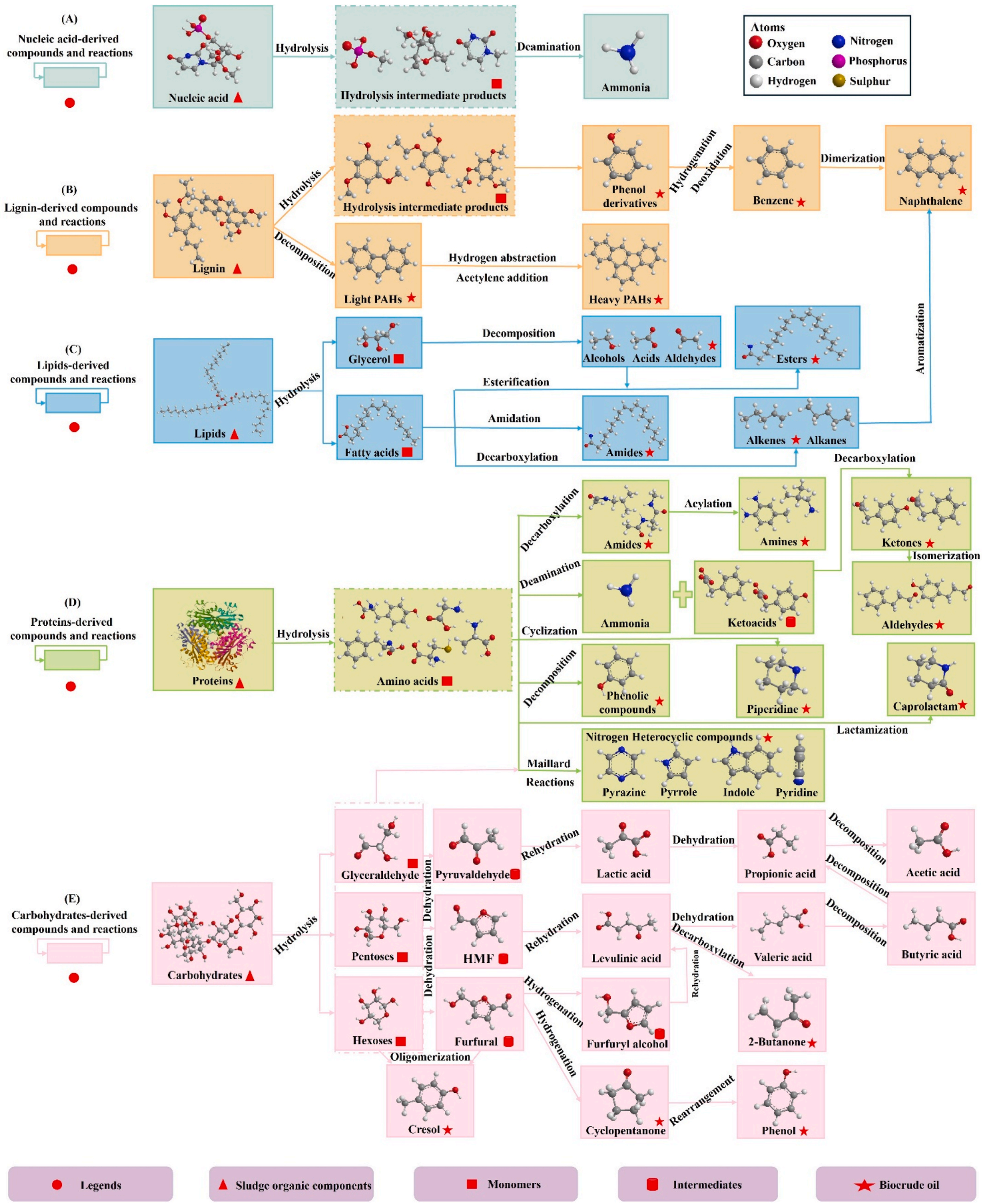

Fig. 6. Potential formation pathways and reaction mechanisms for organic matter conversion in sludge to biocrude oil production through hydrothermal liquefaction. The small squares without geometric shapes indicate that the final products from hydrothermal liquefaction are distributed in the gas or liquid phase.

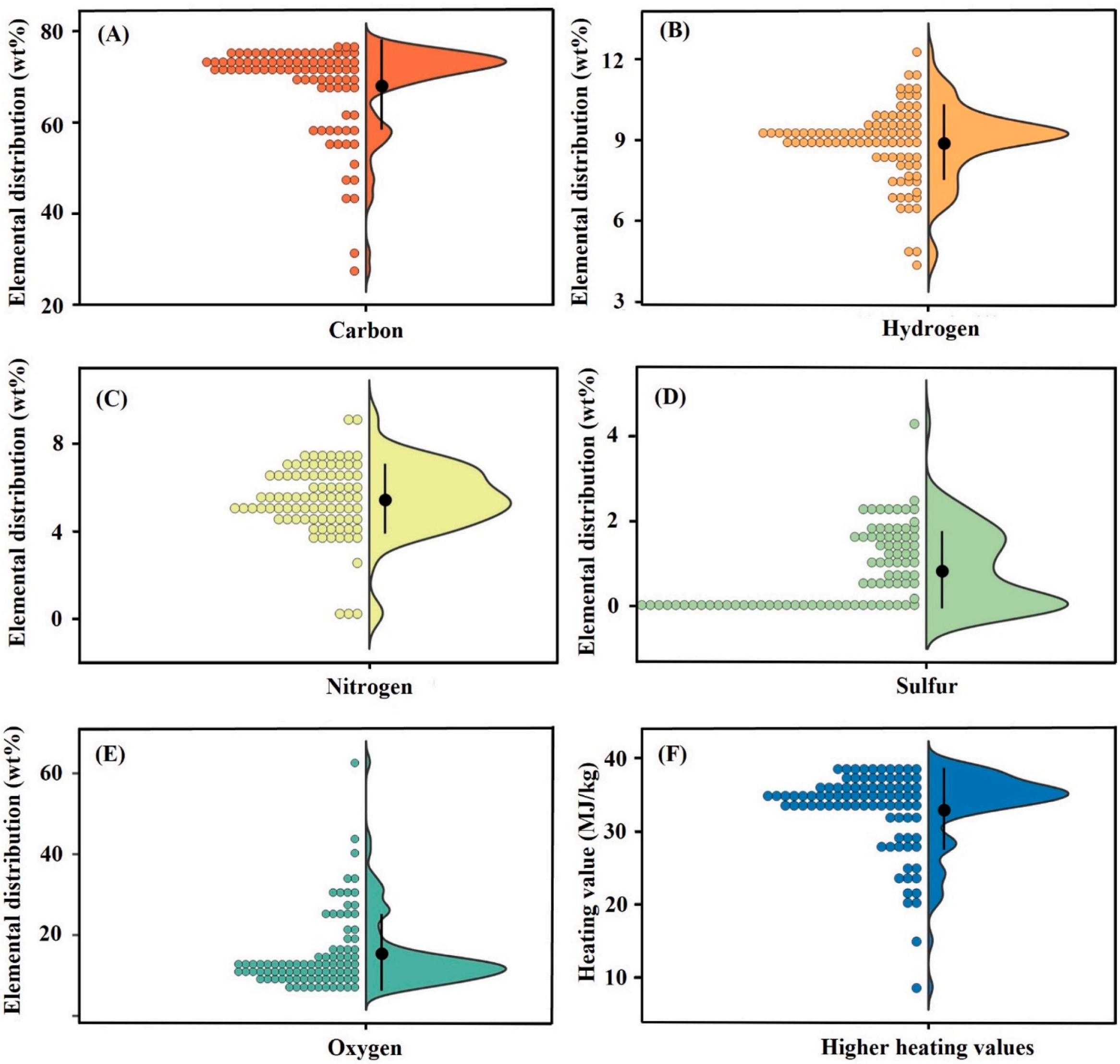

Fig. 7. Violin plot of the recorded elemental components (C, H, N, S, and O) and heating values of biocrude oil from the hydrothermal liquefaction of sewage sludge. Detailed data points are provided in Table S9.

hydrocracking is also efficient in removing heteroatom [96], it is not covered in detail in this review due to its limited usage, warranting further investigation.

Apart from the influence of heteroatoms mentioned above, the quality of biocrude oil derived from the HTL of sewage sludge is often compromised by the presence of water and metals, which can adversely affect subsequent final fuel production (i.e., gasoline, diesel, jet fuel, marine fuel) [93]. Desalting biocrude oil with acids (i.e., formic acid, sulfuric acid) is an effective approach for reducing both water and metal content in raw biocrude oil, eventually facilitating mild hydrotreatment and enhancing the quality of the biocrude oil [97,98]. For instance, washing biocrude oil with formic acid (0.5 % w/w) at 80 °C for 1 h effectively reduced its water content from 7.1 % to 2.4 % and decreased the concentration of alkali metal salts by up to 94 % [93]. Sulfuric acid (0.1 M) wash, achieved a demetalization rate of 89.2 % for sludge-derived biocrude [93]. This effectiveness is primarily attributed to the ease with which metal carboxylates can be demetallized in the presence of acids.

3.4. Fate and distribution of element components in sludge during hydrothermal liquefaction processes

Element such as nitrogen, phosphorus, and heavy metals remarkably influence byproduct (i.e., aqueous phase, solid phase) treatment and the upgrading process of biocrude oil [7,29,30]. Understanding the fate and distribution of these elements is essential for selecting appropriate treatment methods for byproducts and optimizing biocrude oil processing, thereby promoting waste valorization and advancing the development of a circular economy. A detailed discussion of these elements is provided in Section 3.4.1-3.4.3.

3.4.1. Fate and distribution of nitrogen during hydrothermal liquefaction of sewage sludge

Total nitrogen (TN) in sewage sludge consists of both inorganic nitrogen and organic nitrogen (Org-N), with nitrogen primarily connected through nitrogen-carbon bonds or peptide bonds between amino acids in amines and amides [29]. Inorganic nitrogen is mainly present as ammonium-N (NH_4^+-N), the dominant species, while nitrate-N (NO_3^--N) and nitrite-N (NO_2^--N) contribute less than 0.1 % TN [68,99] and Org-N includes compounds such as amino acids and proteins [100]. Previously studies have shown that over 80 % N in raw sludge is transferred to the aqueous phase after HTL, primarily in the form of Org-N [68]. Around 20 % of N is found in hydrochars [100], followed by up to 10 % in biocrude oil, where nitrogen mainly exists as N-heterocyclic compounds [61]. The N content in the gaseous phase is negligible [101].

The possible transformation mechanisms of nitrogen during HTL of sewage sludge are illustrated in Fig. 8. Initially, inorganic N is hydrolyzed to NO_3^--N, NO_2^--N and NH_4^+-N (aqueous phase), while protein-N decomposes into soluble Org-N, which undergoes deamination to form NH_4^+, contributing to gaseous ammonia (NH_3) [100] (Fig. 8). Notably, some NH_4^+-N can convert to NO_3^--N via redox reactions when the temperature exceeds 300 °C (Fig. 8). The N-containing compounds in biocrude oil are classified into three types: amine-N, heterocyclic-N, and nitrile-N (Fig. 8). Amine-N forms through decarboxylation of amino-N and further converts into more stable heterocyclic-N compounds, such as pyrrole-N, pyridine-N, and quaternary-N, via cyclization or Diels-Alder reactions [29,100,102]. Moreover, nitrile-N in the oil phase is produced through alkylamine dehydrogenation [103]. Additionally, N-containing compounds in the solid phase primarily include amino-N, quaternary-N, pyrrole-N, and pyridine-N formed through alkylation, decarboxylation, cyclization, and polymerization, with pyridine-N intermediates sometimes converting to quaternary-N [104]. Amino-N can also react with carbohydrates through Millard reactions, generating nitrogen-containing compounds like pyrrole and pyridine [29], as discussed in Section 3.2.

3.4.2. *Fate and distribution of phosphorus during hydrothermal liquefaction of sewage sludge*

Sewage sludge, containing about 3 wt% phosphorus (P) on a dry weight basis, is considered an important P source, with most of the P retained in hydrochars after HTL [105,106]. The P species in sewage sludge consists of organic P (OP) and inorganic P (IP), non-apatite inorganic P (NAIP), and apatite P (AP) [30], with OP in sewage sludge likely contributing a significant portion of the available P [107]. During the HTL process, P can be immobilized into various species, such as Calcium-phosphate, Iron-phosphate, Magnesium-phosphate, and Aluminum phosphate complexes due to the presence of multivalent metals (i.e., Ca, Fe, Mg, and Al) [30]. Moreover, most OP is converted into IP in hydrochars due to the hydrolysis of organic phosphates and polyphosphates in microbial cells, leading to the release of orthophosphates [108]. Meanwhile, NAIP, primarily base-extractable due to the dominance of iron in the sludge, is the major P fraction in raw sludge, but over 30 % was converted into AP (acid-extractable) after HTL, as calcium phosphates are more thermally stable [30]. Liu et al., observed a remarkable gap between TP and the sum of OP and IP under HTL conditions of 290 °C for 15 min, likely due to the formation of pyrophosphate at lower hydrothermal temperature, which may have been underestimated by the colorimetric method [109]. Speciation results suggest that direct alkaline extraction is effective for recovering NAIP from sewage sludge, whereas acidic extraction is more suitable for phosphorus recovery from hydrochars, which will be discussed in detail in Section 3.5.

3.4.3. *Fate and distribution of heavy metal ions during hydrothermal liquefaction of sewage sludge*

During the HTL of sewage sludge, most heavy metals (i.e., Cu, Zn, Cr, Pb, As, and Cd) concentrated in hydrochars, with less than 10 % ending up in the biocrude oils [7]. For instance, 93.2–98.9 % of Cu, Cr and Zn are retained in biochar [50], predominantly in stable forms, such as residual and oxidizable components [110]. In contrast, 4.5–11.3 % of Zn and Cu are incorporated into biocrude oil [50], attributed to their strong affinity for organic matters and interactions with hydrolysis products of extracellular polymer [111]. Despite the low proportion of heavy metals in biocrude oil, their total concentrations (i.e., 6.7–121.0 mg/kg for Pb, Cu, Zn, and Ni) [7] are remarkably higher than those in regular petrodiesel (0 mg/kg for Pb and Cd, 0.081–0.097 mg/kg for Cu, 0.110–0.141 mg/kg for Zn, 0.005–0.022 mg/kg for Cr, and 0–0.045 mg/kg for Ni) [84,99,112]. These elevated levels suggest potential risks if such biocrude oil are directly used as fuels, as heavy metals could be emitted into atmosphere, posing threats to human health [113]. Moreover, the environmental risks of heavy metals in hydrochars are remarkably reduced compared to raw sewage sludge [7]. For instance, the risks posed by Pb, Cu, and Ni in hydrochar are negligible or low, in contrast to the high risks in raw sludge [7]. Similar findings indicate that HTL effectively decreases pollution levels by transforming mobile heavy metal fractions (i.e., exchangeable/acid soluble and reducible fractions) into stable forms (i.e., residual and oxidizable fractions) [114–116]. However, Zn remains an exception, presenting medium risks even post-liquefaction treatment, although this marks a reduction from the high risk in raw sludge [7]. This suggests that hydrochars, while less hazardous, require further treatment to ensure safe applications.

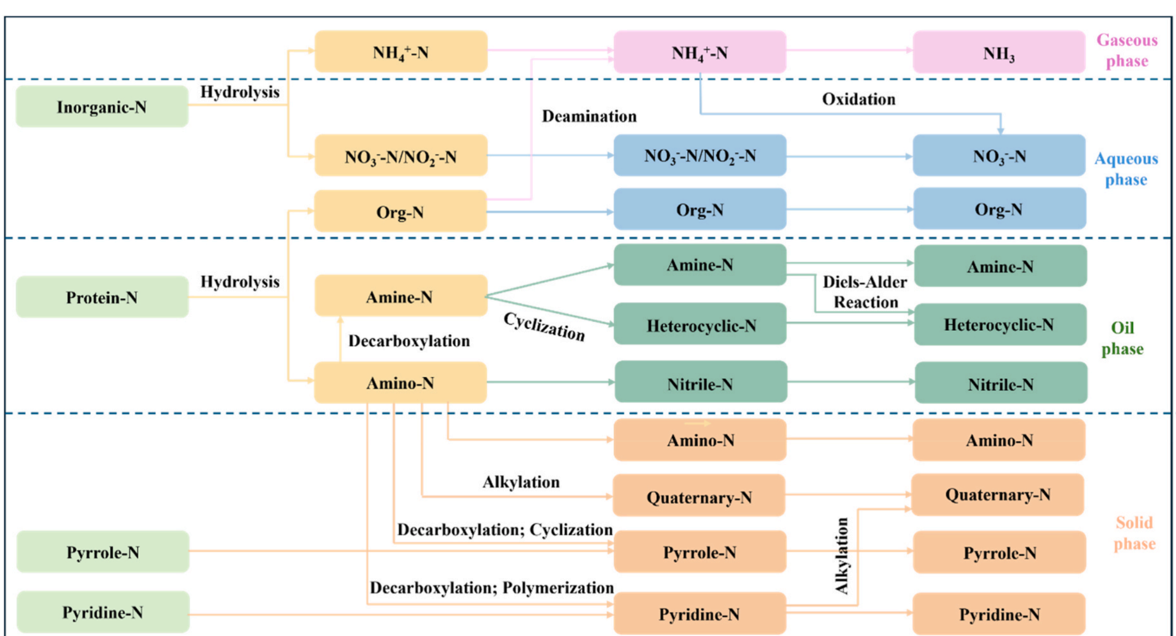

Fig. 8. The possible transformation mechanisms of nitrogen in sewage sludge during hydrothermal liquefaction. Note: N: nitrogen; Org-N: organic-nitrogen.

Additionally, the possible reaction mechanisms of heavy metals during HTL were further elucidated using model compound, with potential reaction pathways illustrated in Fig. 9. Cu incorporation notably increased alkane content by facilitating the hydrogenation of cyclic alkenes and ring-opening reactions, favoring alkane formation over aromatics. However, it also slightly elevated oxygenate concentrations in biocrude oil (Fig. 9A). In contrast, Zn, Cd, and Cr primarily enhanced the content of nitrogen-containing compounds and aromatics (Fig. 9B–D). This effect likely stems from their role in facilitating deoxygenation and dehydrogenation of oxygen-containing heterocyclic compounds as well as promoting Millard reactions between amino and carbonyl compounds. Pb markedly increased ester content-by up to 2.6 times-by enhancing fatty acids formation (Fig. 9E), a trend also observed with As, as reported by Ref. [117]. Collectively, these findings highlight diverse catalytic effects of heavy metals on biocrude oil composition, providing insights into their roles in tailoring oil quality during hydrothermal.

3.5. Possible applications of byproducts from hydrothermal liquefaction of sewage sludge

Biocrude oil, which is the target product of HTL of sewage sludge (~26 wt%), is accompanied by significant byproducts distributed across the solid (22.03 wt%), aqueous (36.67 wt%), and gaseous phases (13.71 wt%) (Fig. 10). These byproducts have considerable potential for various applications, presenting opportunities for resource recovery and nutrient recycling, aligning with the principles of a circular economy. The following sections will provide a comprehensive discussion of these byproducts.

3.5.1. Possible applications of solid phase from hydrothermal liquefaction of sewage sludge

Hydrochars, a solid byproduct of sewage sludge HTL, yield between 0.43 % and 50.73 % on a dry basis, with an average of 22.03 % (Fig. 10), depending on process conditions and sludge properties [3]. The HTL process can mitigate heavy metal mobility and risks [30], yet this may create the misconception that hydrochars are safe for land application. Moreover, long-term use in soil may increase the risk of heavy metals accumulation in the food chain, linked to the leachability and chemical speciation of hazardous constituents [118]. Despite being present in small quantities, hydrochars are rich in valuable resources like phosphorus, metals, and carbon, offering the potential for holistic management, resource recovery, and pollution control [30,106]. Consequently, hydrochars hold significant potential for applications in nutrient and metal recovery, renewable adsorbents, solid fuels, etc. The following sections will explore these applications in detail.

3.5.1.1. Hydrochars used as a promising source of nutrient recovery. Hydrochars are promising sources for phosphorus recovery, contributing to resource recovery and environmental sustainability. Studies report that 85 %–92 % of phosphorus is recovered in hydrochars after HTL of sewage sludge, with total phosphorus content reaching up to 10 % by dry weight [118]. This concentration is comparable to phosphate rock (11 %–15 % phosphorus), with low-grade phosphorus rock typically containing around 8 % [30,119]. These findings suggest that sludge-derived hydrochars could be a valuable source for phosphorus recovery and recycling, addressing environmental challenges and global phosphorus demands [120]. Wet chemical extraction (i.e., acidic extraction) is commonly used for phosphorous recovery due to its

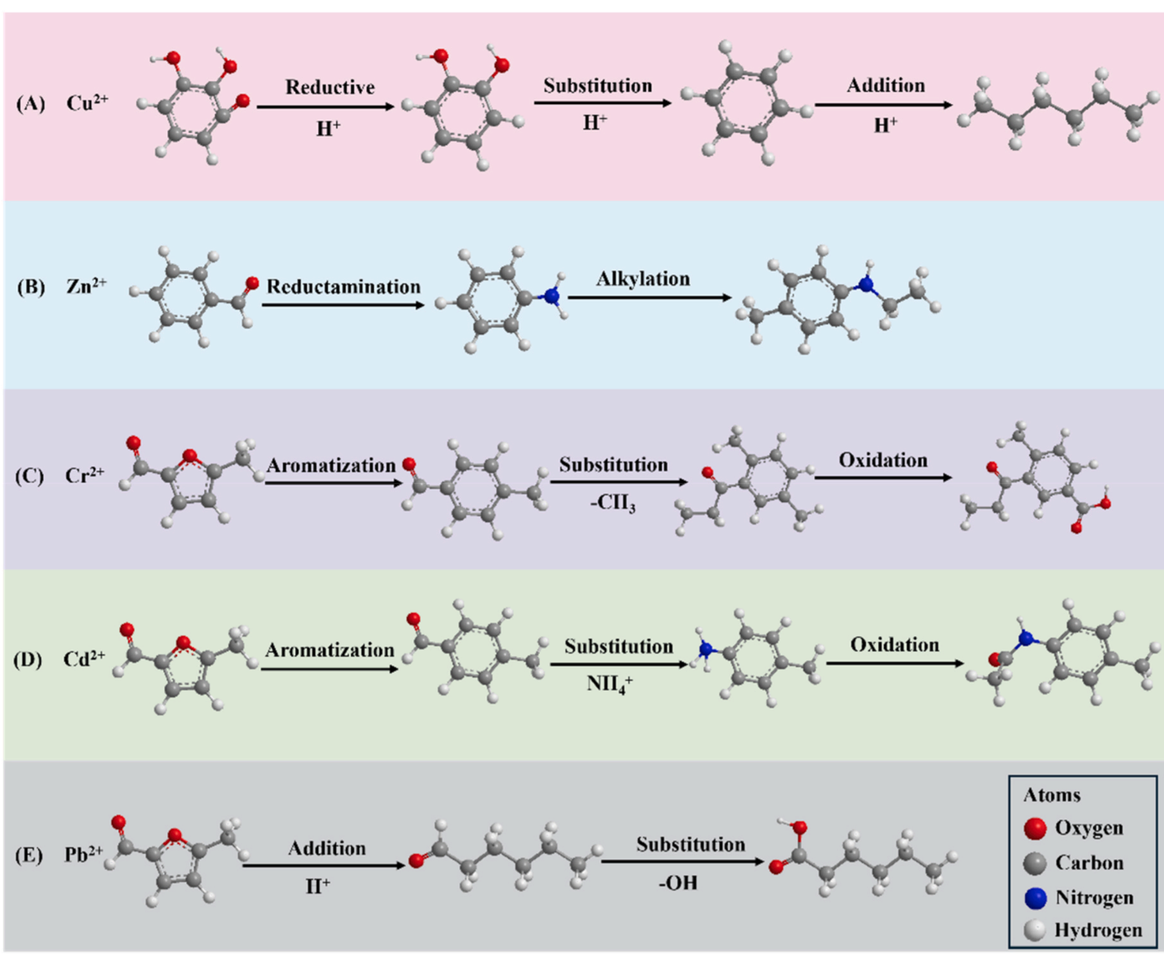

Fig. 9. Possible reaction mechanisms of heavy metals in the production of biocrude oil. Notably, these reaction mechanisms are based on the model compound.

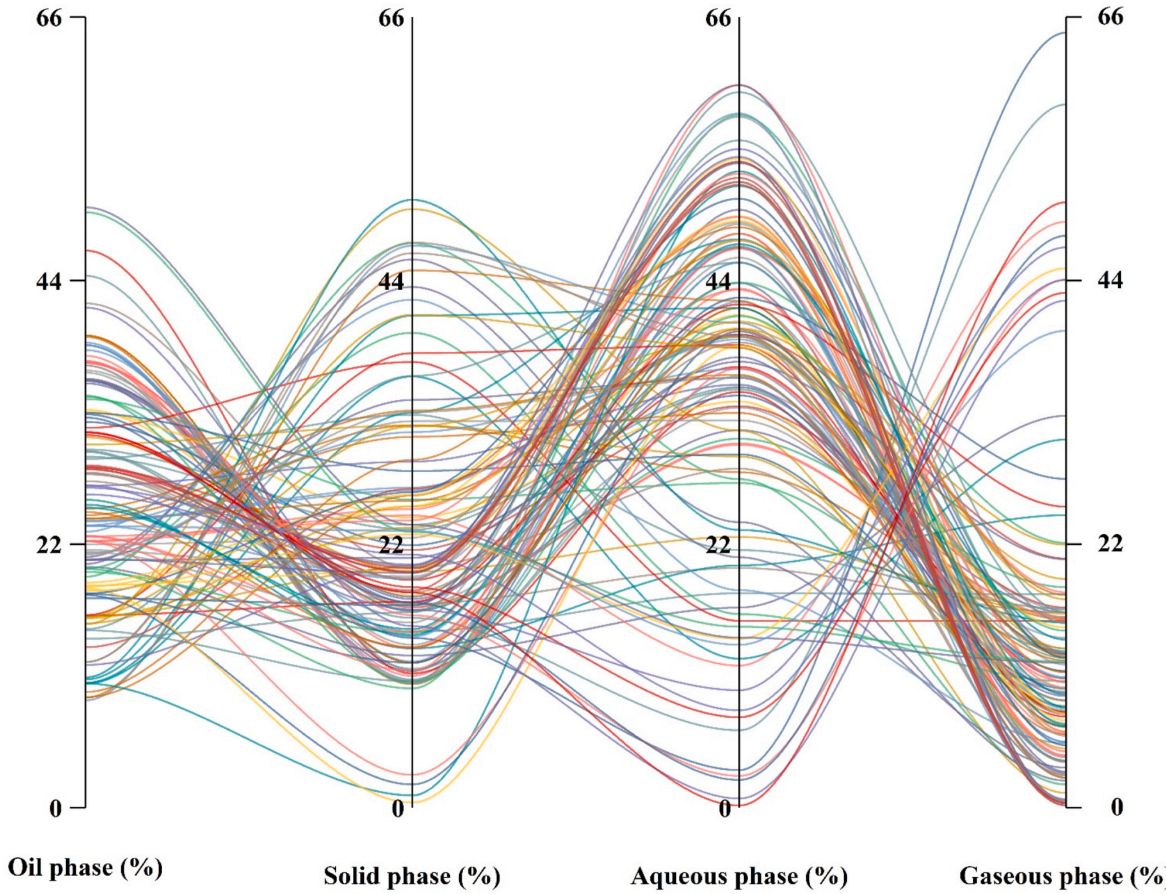

Fig. 10. Distribution of product phases under various conditions during hydrothermal liquefaction of sewage sludge. The colored lines represent individual experimental trial result, showing the relative distribution percentage across the oil, solid, aqueous, and gaseous phases. The highest and lowest points on each phase correspond to the maximum and minimum contribution to that specific phase, respectively. Moreover, the percentages presented in this figure are calculated based on the dry weight of the sludge samples. For the original data used to create this figure, please refer to Table S10 in the Supplementary Materials.

simplicity, high efficiency and cost-effectiveness [120]. For instance, Liu et al., found that the leaching with HNO_3 (0.6 N HNO_3/g for 2 h) achieved near-complete phosphorus extraction. Moreover, Xu et al., showed that H_2SO_4 outperforms HCl, citric acid and acetic acid, achieving 98.7 % efficiency under optimal conditions (0.3 mol/L H_2SO_4, 50 mL/g, 2 h), with a total heavy metals extraction concentration of 30.06 mg/L [121]. In contrast, Pérez et al., found H_2SO_4 and HCl equally effective, with nearly complete phosphorus removal in a 2.5 M acid solution after 2 h, transferring over 70 % of metals like Cu, Fe, Mg, and Zn to the leachate. This variation in phosphorus removal efficiency may be due to differences in sludge properties and HTL conditions, which affect phosphorus and metal concentrations in hydrochars.

Additionally, the extraction mechanisms using the acid extraction is illustrated in Fig. 11. Generally, the phosphorus leaching mechanism can be summarized into four steps: Adsorption (I), Reaction (II), Diffusion (III) and Completion (IV) (Fig. 11). In the adsorption step (I), hydrogen ions (H^+) rapidly distribute themselves on the outer surface of the hydrochars. In the rection (II) step, the acid reacts with surface-bound phosphates. During diffusion (III) step, H^+ ions gradually penetrate the inner surfaces of the hydrochars particles. Finally, in the completion (IV) step, the reactions proceed deeper into the particle cores until complete. Notably, the dissolution of calcium-phosphate minerals plays a critical role in the phosphorus extraction process from hydrochars produced through the HTL of sewage sludge [106].

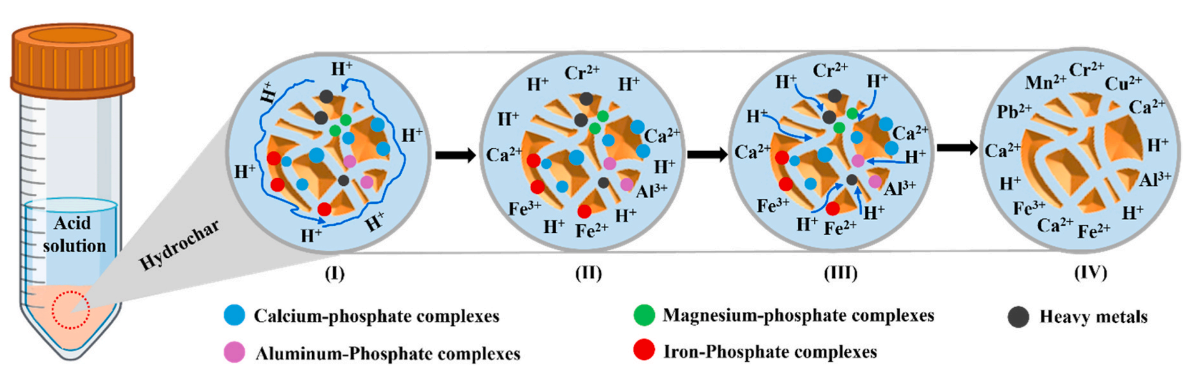

Fig. 11. Phosphorus leaching processes from hydrochar, a byproduct of the hydrothermal liquefaction of sewage sludge, using acid extraction techniques.

3.5.1.2. Hydrochars used as a renewable adsorbent. Hydrochars are regarded as a renewable alternative to fossil-derived adsorbents, offering sustainable solutions for adsorption applications. Its adsorption capability is primarily attributed to oxygen-rich functional groups, porous surface structure, and thermal stability, making it highly effective for various adsorption processes [122,123]. Leng et al., showed that hydrochars removed up to 144.2 mg/g of methylene blue, surpassing pyrolytic hydrochars (12–130 mg/g) [124,125], likely via monolayer chemisorption. Marx and van der Merwe demonstrated that hydrochars from paper sludge effectively removed organic pollutants, such as phenol and its derivatives, from both synthetic and industrial wastewater, reducing phenol levels from 3.2 mg/L to<0.003 mg/L, indicating near-complete removal [126]. Interestingly, demineralization was found to significantly enhance the adsorption performance [127]. For example, methylene blue adsorption capacities reached 87.6, 297.4, and 367.1 mg/g for raw, acid-washed, and demineralized hydrochars, respectively, compared to 332.3 mg/g for commercial-activated carbon [127]. Acid washing altered the adsorption mechanism from Langmuir-monolayer adsorption to Freundlich-multilayer adsorption [127]. Both demineralized hydrochars (with and without acid washing) and commercial activated charcoal were found to be highly effective in removing pharmaceuticals, which are typical organic micropollutants from real wastewater treatment effluents, achieving waste sustainable management, contributing to circular economy development. These findings suggest that residues from nutrient extraction (i.e., acid leaching) in HTL can serve as renewable adsorbents.

3.5.1.3. Hydrochars used as a solid fuel. Hydrochars from sewage sludge HTL typically have ash content exceeding 45 % (up to 88 %) and low calorific values (<10 MJ/kg), making them less suitable as untreated fuels compared to hydrochars from hydrothermal carbonization [29, 30]. Hydrochars can serve as solid fuel after pretreatment, such as acidic extraction, which enhances combustion performance, while direct burning is discouraged due to these limitations. High ash content and an alkali index of 0.28–0.72 kg/GJ pose slagging and fouling risks during composition, as direct burning can lead to the formation of alkali sulfates and silicates that deposit on combustor surfaces [30,128]. Acid modification effectively mitigates these risks by removing alkaline earth and alkali metals while improving fuel properties [118,129]. Liu et al., found that HNO_3 leaching (0.6 N HNO_3/g, 2 h) reduced ash content to 34 %, increased carbon content to 48 %, and raised the heating value to 20.5 MJ/kg, compared to raw hydrochars. Additionally, combustion occurs in three stages: moisture evaporation (30–150 °C), followed by devolatilization, or volatile combustion (150–350 °C), and finally, fixed carbon or char combustion (350–550 °C). Acid modification did not alter these mechanisms but reduced activation energy during the initial stage, providing a suitable pathway to transform hydrochars into solid fuels [118]. Therefore, it is worth investigating whether acid modification can enhance the suitability of hydrochars for combustion and support closed-loop recycling.

3.5.2. Possible applications of gaseous phase from hydrothermal liquefaction of sewage sludge

The gaseous phase produced during the HTL of sewage sludge accounts for a significantly smaller proportion compared to other phases ($p < 0.0001$), ranging from 0.2 % to 64.68 %, with an average value of 13.71 ± 12.83 % (Fig. 10). The produced gases consist of CO_2, H_2S, H_2, N_2, CH_4 and CnHm (i.e., C_2H_4, C_2H_6, and C_3H_8, etc.), which are presumably formed via complex reactions, including decarboxylation, pyrolysis, decomposition, thermal cracking, methanization, denitrogenation, water-gas shift, and deamination [80,130,131]. Notably, CO_2 is the predominant component of the gaseous phase from HTL, constituting over 90 % of total gaseous production [36,131]. Due to its low overall proportion and high CO_2 content, many researchers advocate for its direct release into the atmosphere without any treatment [11,132]. However, large-scale implementation of HTL in wastewater treatment plants could generate significant gas emissions, potentially exacerbating the greenhouse effect. Therefore, these emissions warrant treatment rather than direct release. The CO_2-rich gas offers valorization opportunities, such as electrochemical reduction to formic acid, which can serve as an additive during the HTL step [133]. Additionally, the presence of flammable gases accounting for 3.7–9.6 %, highlights the need to consider their collection, treatment, and reuse [131]. Implementing these strategies can support the complete valorization of municipal waste, aligning with sustainable waste management goals.

3.5.3. Possible applications of the aqueous phase from hydrothermal liquefaction of sewage sludge

The aqueous phase generated during the HTL of sewage sludge constitutes the largest proportion among all phases ($p < 0.0001$), ranging from 0.19 % to 60.3 %, with an average of 36.67 ± 14.44 % (Fig. 10). The aqueous phase typically contains 20 %–40 % of the organics (i.e., organic carbon>10 g/L) and 60 %–80 % of the nutrients (i.e., total nitrogen>2 g/L), rendering it unsuitable for direct recycling into conventional wastewater treatment processes [134–136]. If discharged untreated, it poses severe environmental risks, such as eutrophication, nutrient runoff, and harmful effects on water quality due to its high COD (chemical oxygen demand), total organic carbon, and ammonia [137]. Addressing these challenges is critical for scaling HTL technology in wastewater treatment plants, necessitating innovative approaches to valorize the carbon and optimize resource recovery [31,138]. Various methods, including anaerobic digestion, aerobic treatment, recycling, microalgae cultivation, electrochemical oxidation, and wet oxidation, have been explored to manage the aqueous phase effectively.

3.5.3.1. Anaerobic digestion, aerobic treatment, and recycling. Anaerobic digestion is commonly employed for the treatment of aqueous phase but is hindered by inhibitory compounds such as phenolics and ammonia [134,139]. Wang et al., reported complete methane inhibition when using untreated aqueous phase. However, pretreatment methods like struvite precipitation and biochar adsorption restored methane yields to 32.14 and 28.85 mL/g COD, respectively, while a combined approach achieved 225 mL/g COD. These methods are cost-effective, with biochar being inexpensive and struvite offering value as a slow-release fertilizer. Contrarily, Chen et al., achieved methane yields of 136–286 mL/g COD with the use of aqueous phase without pretreatment, suggesting that sludge properties and HTL conditions remarkably influence outcomes. Moreover, conductive materials, such as activated carbon and magnetite, further enhance anaerobic digestion by promoting interspecies electron transfer and enhancing inhibitory substance degradation, as evidenced by methane yield increase of 28.2 % and 25.5 %, respectively [139,141].

Aerobic treatment effectively resists inhibitory compounds such as N-heterocyclics, ammonia, phenolics, furans, and cyclic compounds, while simultaneously addressing nitrogen and phosphorus removal [31, 140]. Kulikova et al., observed that aerobic treatment on the aqueous phase, achieving organic substance removal rates of 67–95 %. However, blending the aqueous phase with influent wastewater raised BOD from 87.1 to 120 mg/L effluent COD from 148 to 172 mg/L, increasing aeration energy consumption by 38 % [142]. Similarly, Liu et al., reported COD increases of 16.3 % and 20.5 % under average and low flow conditions, respectively. UV disinfection efficiency dropped by 4 % and 8 %, likely due to higher UV adsorption from melanoidins and phenolic compounds in the aqueous phase [143]. Additionally, recycling the aqueous phase into HTL units is another approach, but its high nitrogen content (exceeding 50 %) can degrade biocrude oil quality and increase hydrogen demands for upgrading [83]. Activated carbon pretreatment mitigates this issue, achieving up to 73 % nitrogen removal and improving energy recovery from biocrude oil by 11 % [83]. This strategy enhances the sustainability of HTL of sewage sludge as a stand-alone process, as it does not require integration with other treatment processes.

3.5.3.2. Microalgae cultivation. Microalgae cultivation facilitates nutrient removal and recycling from the aqueous phase, supporting sustainable sludge management and a circular economy [16]. The aqueous phase from HTL, rich in phosphorus (0.5–18.9 g/L) and nitrogen (1.9–12.7 g/L), is linked to feedstock protein content and serves as a nutrient source for microalgae [144–146]. Das et al., revealed that *Picochlorum* sp. (microalgae) efficiently utilized nutrients from the aqueous phase, achieving nitrogen removal rates of 95.4 % and higher biomass yields compared to control cultures. In contrast, *Chlorella* sp. showed lower nitrogen removal rates (58.6 %) and reduced biomass yield, likely due to organic compounds in the aqueous phase that inhibited growth. Similarly, Kumar et al., observed reduced biomass yields across four oleaginous algae strains treated with the aqueous phase, attributing this to high COD levels and the presence of TiO_2 catalyst that interfered with intracellular metabolism [147]. These findings, consistent with Xia and Murphy, emphasize that nutrient recycling efficiency is strain-dependent [148]. Notably, stress conditions (i.e., nitrogen limitation) can induce oxidative stress, promoting lipid accumulation through reactive oxygen species formation, and ultimately enhancing lipid productivity [149,150].

3.5.3.3. Wet oxidation, electrochemical oxidation and hydrothermal gasification. Wet oxidation, using oxygen or air as an oxidizing agent, is a promising technology for reducing COD and simplifying the aqueous phase from HTL [137,151]. Thomsen et al., reported total organic carbon and COD removal rates of 96.1 % and 97.6 %, respectively, at 350 °C with a 3 h holding time in a non-catalytic process, which also doubled acetic acid content, creating opportunities for downstream applications, such as microbial electrolysis or biological processes to utilize volatile fatty acids while addressing toxicity concerns [137,152]. Kilgore et al., found catalytic wet oxidation (i.e., WO_3, and ZeO_2) increased acetic acid production by 10–20 % compared to non-catalytic methods. Notably, wet oxidation can generate enough heat to become self-sustaining with effective recovery, as shown in pilot-scale HTL using a heat exchanger that recovers 75 % of the heat [153,154]. However, high-pressure oxygen or air imposes additional energy costs, wet oxidation is more energy-efficient than alternatives such as electrochemical oxidation (28 kWh/kg COD) [155] or urban wastewater treatment plant (0.03–7.1 kWh/kg COD) [156].

The electrochemical oxidation process offers several advantages, including the effective removes refractory inorganic and organic compounds, owing to its simplicity and robustness [157]. Ciarlini et al., reported its efficiency in reducing soluble organic matter in the aqueous phase derived from HTL, but its high energy consumption remains a challenge [158]. An integrated approach combining anodic oxidation with hydrogen production from cathodic reactions has been proposed to enhance economic viability, as illustrated in Fig. 12. Matayeva and Biller demonstrated this method, achieving up to 99 % COD removal and hydrogen production rates of 1.8 NL/h, highlighting its potential for sustainable treatment [159]. Alternatively, catalytic hydrothermal gasification is another method to treat the aqueous phase from HTL by converting organic matter into methane and carbon dioxide [160,161]. However, high capital costs and sulfur poisoning of precious metal catalysts limit its broader application [162].

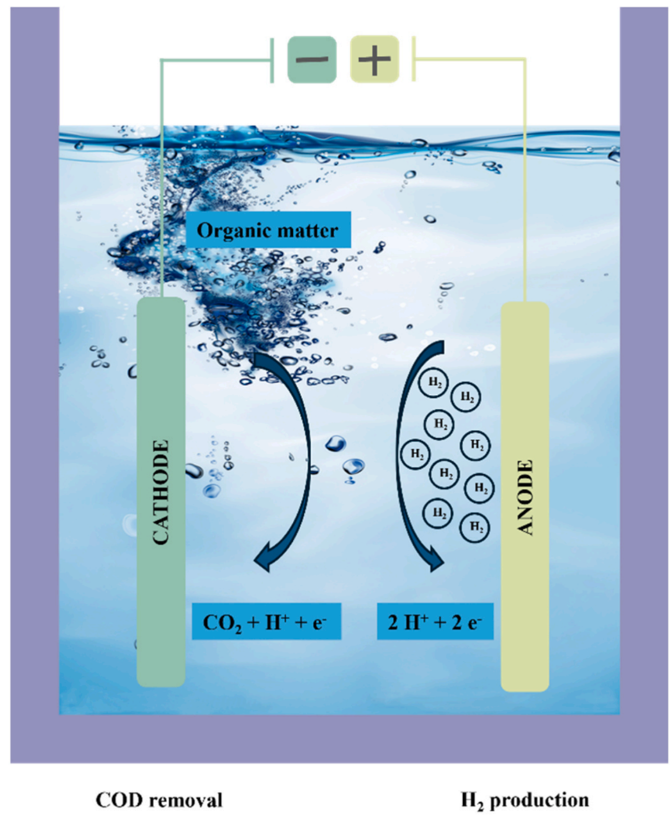

Fig. 12. Schematic diagram of electrochemical oxidation process for treating the aqueous phase from hydrothermal liquefaction of sewage sludge.

3.6. Future perspectives

Previous studies have identified sludge types, operating parameters, catalyst use, and sludge pretreatment as crucial factors influencing biocrude oil production via HTL of sewage sludge [13,15–17]. Mixed sludge is highly recommended for HTL due to its high biocrude oil yields compared to primary and secondary sludge [13,14,41]. Ethanol-water co-solvents show promise for improving efficiency [56], future studies should focus on optimizing solvent usage, developing efficient solvent recovery approach, and evaluating large-scale economic feasibility to enhance both environmental and economic sustainability. Centrifugation outperforms liquid-liquid extraction in lab-scale tests [43], yet scaling up requires further validation, along with the development of eco-friendly alternatives to traditional solvents like dichloromethane. Pretreament methods, such as microwave and chemical treatments, enhance sludge disruption, and increase the availability of organic matter for biocrude oil production, but are energy- and cost-intensive. Waste-derived substances like urine and lignosulfonate offer sustainable alternatives for achieving effective sludge disruption [163,164], though their feasibility and impact on biocrude quality require further investigation. To advance low-cost, eco-friendly methods, future research could explore the use of waste heat integration for pretreatment of sewage sludge, bio-based or recyclable solvents, and low-energy mechanical disintegration techniques (i.e., hydrodynamic cavitation). Additionally, assessing the long-term economic viability and scalability of these strategies remains critical for the sustainable development of HTL. Notably, in typical wastewater treatment plants, the generation of sewage sludge can vary remarkably over time [14]. Consequently, studies that analyze sludge from a single time point are inadequate for assessing the real-world viability of HTL. Future research should prioritize the long-term performance of HTL by simulating fluctuations in feedstock and variations in product characteristics over time.

The potential transformation trends of organic matter in sewage sludge during HTL for biocrude oil production generally follow the order: lipids, proteins, carbohydrates, lignin and/or humic substances [26,72,73]. However, these trends, based on model substrate simulations, do not fully account for the complex and heterogeneous nature of actual sewage sludge. Future research should focus on studying these transformation pathways under real sludge, considering sludge's diverse composition, and exploring interactions between organic fractions to optimize biocrude oil yield and quality. Additionally, the HTL process elevates nitrogen and oxygen content, raising N/C and O/C ratios, with average values of 0.083 and 0.23 (Fig. S4), which are significantly higher than those of petroleum crude [59]. Therefore, future studies should not only aim to maximize biocrude yield but also control heteroatom content to improve biocrude quality and reduce the need for subsequent upgrading, thus enhancing the economic and environmental viability of biocrude production.

The commercialization of biocrude oil is hindered by its quality, primarily due to the presence of heteroatoms and alkali metal salts [92,93]. Oxygen removal from biocrude oil during hydrotreating is effective, but nitrogen removal remains challenging, particularly in biocrude oil with high nitrogen content, impacting the hydrotreating process and fuel quality [92]. Improving nitrogen removal requires tailored catalysts and optimized HTL conditions to minimize heterocyclic compound formation. However, conventional solid catalysts enhance activity but increase coke deposition and catalyst deactivation [96], a challenge mitigated by unsupported dispersed catalysts. Moreover, catalysts tend to accumulate Fe, Ca, Si, and Cr, likely resulting from the erosion of stainless-steel components during hydrotreatment. These findings highlight the importance of developing effective catalysts for these target processes, as well as carefully selecting appropriate conditions and construction materials for further studies on biocrude oil derived from sewage sludge [86]. Additionally, washing the oil phase with dilute acids to remove alkali metal salts remarkably improves biocrude oil quality and reduces hydrogen consumption during subsequent hydrotreating [93]. We proposed an upgrading pathway for biocrude oil with high nitrogen content derived from sewage sludge, as shown in Fig. 13. The pathways involve pretreatment through desalting followed by two-stage catalytic upgrading-oil stabilization in the first stage and fuel quality enhancement in the second stage [95]. Desalted biocrude oil offers advantages, such as extended catalyst lifetimes and increased oil yields [93]. The upgraded oil is distilled to produce target products, like gasoline (<150 °C), jet fuel (150–250 °C), marine fuel (250–360 °C), and asphalt (>360 °C) [165]. However, while dilute acid washing is effective, it may lead to oil loss and higher operational costs [166], necessitating a comprehensive life cycle assessment to evaluate economic feasibility. Future research should also explore novel refining technologies to further enhance furl performance and quality.

Solid phase, byproduct of HTL, holds potential for nutrient recovery (i.e., phosphorus and metal ions) and solid fuel production [30,119]. However, several critical areas require further investigation to optimize this potential, mainly including: 1) assessing the long-term stability of phosphorus in extracted hydrochars is essential for determining their feasibility of reuse after recovery; 2) evaluating the environmental implications (i.e., land applications) of phosphorus recovery to ensure that they do not inadvertently harm ecosystems or human health. For assessing the potential of hydrochars as solid fuel, more comprehensive investigations are needed to determine if desirable fuel can be produced post-phosphorus recovery. Understanding how acid modification affects hydrochars characteristics and combustion performance is essential. Additionally, aqueous phase presents challenges for treatment due to inhibitory substances like furans, N-heterocyclics, and phenolics [139].

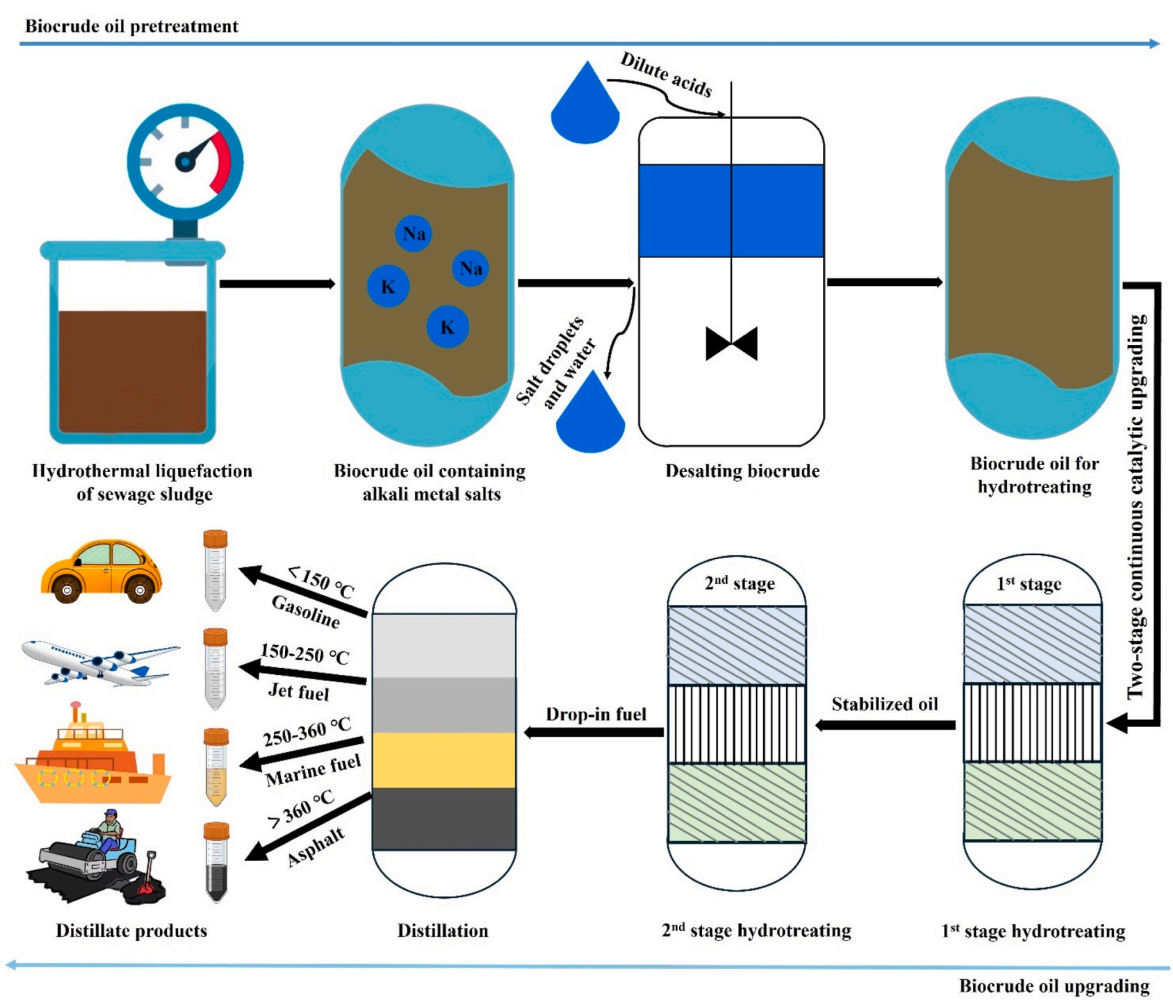

Fig. 13. Proposed upgrading pathways for biocrude oil with high nitrogen content derived from the hydrothermal liquefaction of sewage sludge.

While anaerobic digestion has shown potential for effectively treating this phase, most studies have been limited to batch-scale experiments. To develop a robust and scalable anaerobic digestion process, further research using continuous-flow reactor is essential. For the aqueous phase treated via aerobic processes, future studies should prioritize isolating microbial communities capable of effectively degrading recalcitrant compounds and optimizing conditions for preliminary physicochemical treatment of wastewater. These advancements will enhance the environmental safety and commercial viability of the HTL process.

4. Conclusions

This study offers the first comprehensive and systematic analysis of biocrude oil production from the HTL of sewage sludge. The findings provide valuable references and provide clear guidance for advancing HTL technology toward industrial applications. The key insights from this review are outlined below:

1) Mixed sludge, with its balanced organic matter composition, is ideal for biocrude production, achieving an average yield of 38.95 % (range: 35.3–42.6 %). Reaction temperature is more critical than holding time in biocrude oil production, with higher biocrude oil yields more likely to be achieved at approximately 350 °C and 30 min. Ethanol-water co-solvents as the reaction medium have better performance compared to sole solvent, and centrifuge extraction proves more effective than liquid-liquid extraction methods. Long-term, pilot-scale, and full-scale studies, along with life cycle assessments, are essential to validate these findings and evaluate their economic feasibility.
2) Organic matter transformation for biocrude oil yields follows the trends: lipids > proteins > carbohydrates > lignin or humic substances. The formation of biocrude oil involves complex reactions, including hydrolysis, decomposition, deamination esterification, amidation, decarboxylation, dimerization, rearrangement, depolymerization, etc.
3) Biocrude oil contains significant amounts of heteroatoms (average: N: 5.5 %, S: 0.9 %, and O: 15.7 %), making their removal essential for upgrading to meet biocrude oil applications, with hydrotreating becoming a promising method for this process. Moreover, the presence of metals and water also remarkably reduces biocrude oil quality, but desalting with acids can mitigate this issue.
4) The nitrogen component primarily distributes into the aqueous phase during HTL, while phosphorus and metal ions predominantly transfer into the solid phases, providing potential for further resource recovery of nitrogen, phosphorus, and metals.
5) The HTL process not only produces biocrude oil but also generates byproducts predominantly in the aqueous phase (36.67 %), followed by the solid phase (22.03 %) and gaseous phases (13.71 %). The aqueous phase can be managed via anaerobic digestion, aerobic treatment, microalgae cultivation, while the solid phases offer potential for nutrient and metal recovery, renewable adsorbents, and solid fuels, and the gaseous enables valorization through processes like electrochemical reduction to formic acid.

CRediT authorship contribution statement

Zhenyao Wang: Writing – original draft, Investigation, Visualization, Conceptualization, Formal analysis, Writing – review & editing. **Xuan Li:** Supervision, Visualization, Formal analysis, Writing – review & editing. **Huan Liu:** Writing – review & editing. **Carol Sze Ki Lin:** Writing – review & editing. **Qilin Wang:** Supervision, Resources, Funding acquisition, 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.

Acknowledgements

Qilin Wang acknowledges Australian Research Council (ARC) Future Fellowship (FT200100264).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2025.116086.

Data availability

Data will be made available on request.

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