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Review

Life Cycle Assessment of Hydrothermal Carbonization: A Review of Product Valorization Pathways

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Abstract: Hydrothermal carbonization (HTC) has the potential to be a sustainable and environmentally beneficial approach for organic waste treatment. It is likely that HTC product use will dictate the viability of large-scale HTC facilities; therefore, understanding the viability and environmental implications associated with HTC product valorization pathways is critical. The overall goal of this review is to gain an understanding of how HTC product valorization is currently being modeled in life cycle assessment studies, and to use such information to assess current research and/or data needs associated with product valorization. To accomplish this, a review of existing HTC literature was conducted and used to assess the current state of knowledge surrounding the environmental implications of HTC product use. From this review of the literature, it is clear that potential exists for HTC product valorization. To realize this potential in a full-scale application, research gaps and data needs were identified that included a system-level integration to evaluate location-specific information as well as more extensive characterization of the impact of HTC product properties on valorization impacts.

Keywords: hydrothermal carbonization; life cycle assessment; product valorization; hydrochar; process water; review; environmental implications

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1. Introduction

Hydrothermal carbonization (HTC) is a thermal conversion process that has been gaining significant interest as a sustainable and environmentally beneficial way to manage a variety of organic waste types, including sewage sludge [1], municipal solid wastes (MSW) [2], food waste [3], industrial biomass [4,5], lignocelluloses and microalgal biomass [6], and livestock manures [7]. HTC processes occur at relatively low temperatures (180–250 °C) under autogenic pressures. During HTC, the feedstock is decomposed by reaction mechanisms similar to those in dry pyrolysis, which include hydrolysis, dehydration, decarboxylation, aromatization, and recondensation [8,9]. As a result, wet biowaste is converted to three value-added products: (1) a condensed carbon and energy-rich solid material known as hydrochar (HC); (2) a liquid phase, often referred to as process water (PW), which is rich in organic carbon and nutrients; and (3) a gas-phase that is mostly comprised of CO_2, but also contains some energy-rich gases. HC has considerable valorization potential, lending itself to numerous applications such as a soil amendment [9], solid fuel [9–11], adsorption media [12–14], and for soil remediation [15]. Despite the fact that the PW retains a substantial amount of carbon and nutrient content, only a small number of studies have investigated PW valorization. Leng et al. [16] reviewed various PW valorization pathways, including using it as a nutrient source for microalgae growth, nutrient recovery, conversion of organics to biofuels, and recovery of valuable compounds. Even fewer studies have explored volatilization pathways for the gas-phase, which retains a much smaller percentage of carbon but contains some energy-rich gases [2,3].

It is likely that HTC product use and their associated markets will dictate the feasibility of large-scale HTC facilities. Thus, understanding the viability and environmental implications associated with HTC product valorization pathways is critical. It is also important that policies associated with HTC product use be developed. The proportion of studies demonstrating effectiveness of HC as a competent contender to substitute fossil fuel coal in energy production has encouraged the publication of a few policy-related studies [17]. Gaining insight into the level of environmental impact linked to specific combinations of HTC feedstocks and product applications can be used to guide the optimization of process conditions and ultimately inform the development of relevant policies.

Life Cycle Assessment (LCA) is a technique that has been used to evaluate the potential environmental impacts of the HTC process [18,19]. LCA is a technique widely used to assess and quantify the potential environmental impacts of a system or service throughout its entire life (e.g., cradle to grave), and includes all relevant inputs from natural systems (e.g., crude oil, iron ore, water) and emissions (e.g., carbon dioxide, methane) to different components of the environment (e.g., soil, water, air), ultimately linking them to potential environmental and human health impacts (e.g., global warming or toxicity). Recent review articles have detailed aspects of HTC-related LCA studies [20–23]. The majority of these studies conducted to date compare HTC with other waste management processes, such as anaerobic digestion (AD), composting, and incineration, as has been reported by Sarrion et al. [24]. An important aspect of these studies is the modeling of HTC product valorization and the influence such valorization may play on overall system impact. LCA may serve as a quantitative indicator of long-term environmental benefits and burdens that shape guidance related to how, when, and where HTC product valorization should be conducted. LCA results can also be used to develop environmental policies [25].

The overall goal of this review is to gain an understanding of how HTC product valorization is currently being modeled in LCA studies and to use such information to assess current research and/or data needs associated with product valorization. To accomplish this, a review of existing HTC literature was conducted and used to assess the current state of knowledge surrounding the environmental implications of HTC product use. Results from the review of the literature were used to identify research gaps and needs associated with HTC product valorization. Such information can ultimately be used to guide the development of policies and regulations associated with HTC product valorization.

2. Materials and Methods

The goal of this review is to gain an understanding of how HTC product valorization is currently being modeled in LCA studies and to use such information to assess current research and/or data needs associated with product valorization. To collect studies to meet this goal, a combination of Google Scholar and ScienceDirect databases were used. Collected studies were limited to peer-reviewed journal articles in which "LCA of HTC" was found in the title, abstract, or as an author-specified keyword. Different combinations of these keywords were used to ensure all applicable studies were collected. When applying these restrictions, a total of 46 peer-reviewed articles were identified. The publications reviewed in this study are from database search results of articles published as of 14 November 2023. Because the goal was to review studies in which an LCA was conducted, papers included in this effort needed to have performed a full scope of an LCA that included: (1) a functional unit, (2) a clearly defined system boundary used for analysis, and (3) at least one impact category (e.g., global warming potential (GWP)). This additional criterion reduced the total applicable studies reviewed for this specific effort to 34 papers. When collecting data from these studies, specific details associated with the LCAs surrounding the HTC process and/or results from comparisons between HTC and other processes were not used. Instead, data collection was focused on how the environmental implications of the HTC-generated products were modeled.

3. Results

3.1. Overview of Collected Studies

A total of 34 papers associated with the LCA of organics that met the criteria for inclusion in this effort were collected and reviewed. A summary of these studies is included in Figures 1 and 2, which illustrate the connection between feedstock, valorization pathway, life cycle assessment impact assessment (LCIA) method, and number of impact categories used in the assessment. Table 1 provides more specific detailed information about each study, including functional units and study goals. Trends associated with these collected studies are illustrated in Figure 3. As shown, conducting LCA studies on HTC processes is a growing field, with the number of publications generally increasing from 2015 to 2023. It should be noted that the number of these studies is quite small in comparison with the yearly publications associated with other aspects of the HTC process. Additionally, these collected studies represent efforts from research groups around the world (Figure 3b) and focus on a variety of feedstocks (Figure 3c). Food waste is the feedstock in which the majority of LCA papers focused, with sewage sludge and the organic fraction of MSW also being well studied. These specific feedstocks have been the focus of significant fractions of other HTC-related publications, so this observation is not surprising.

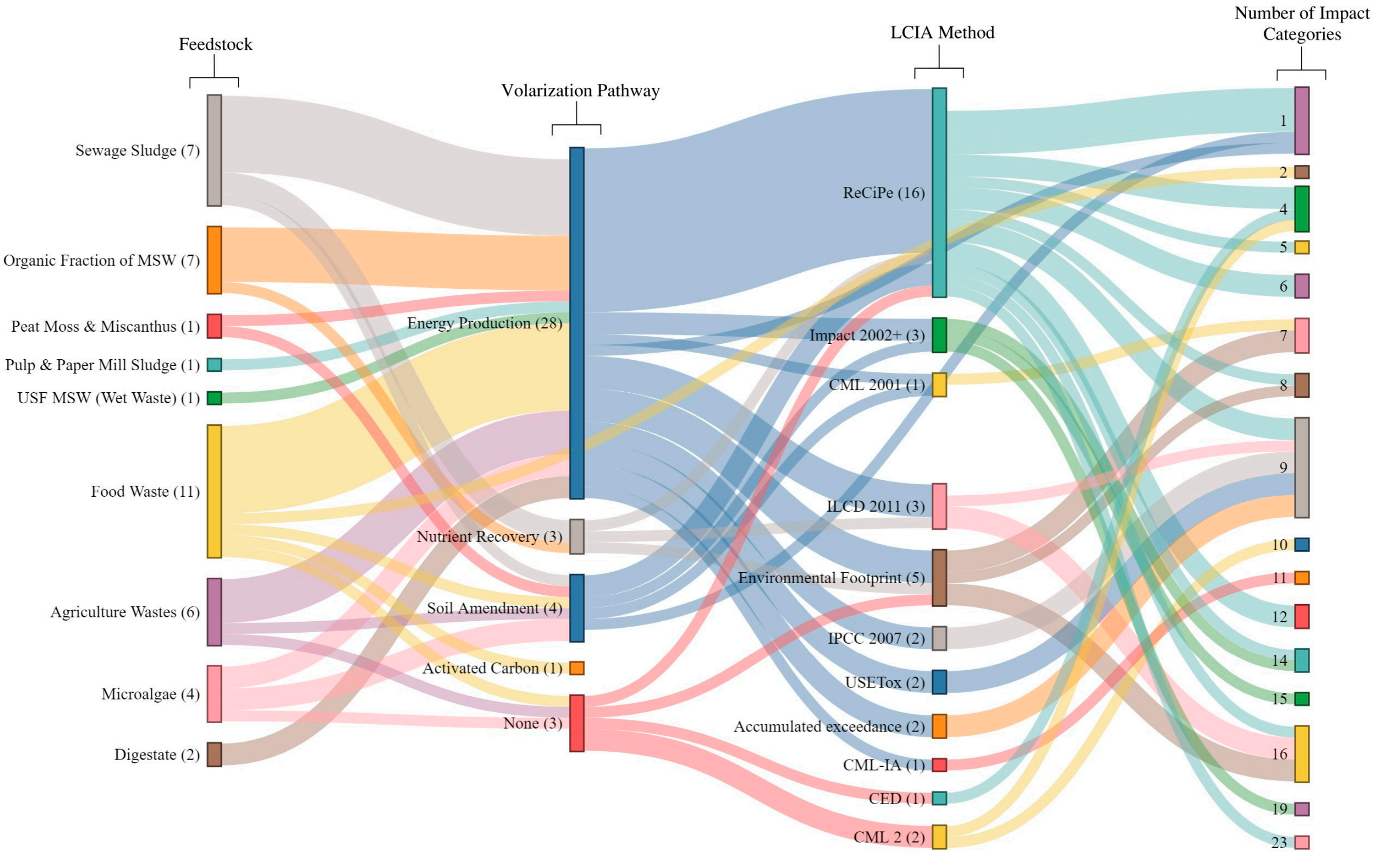

Figure 1. Sankey diagrams illustrating the connection between feedstock, valorization pathway, LCIA method, and number of impact categories for the collected studies for the hydrochar. The numbers in parentheses represent the number of collected studies associated with each category in the figure.

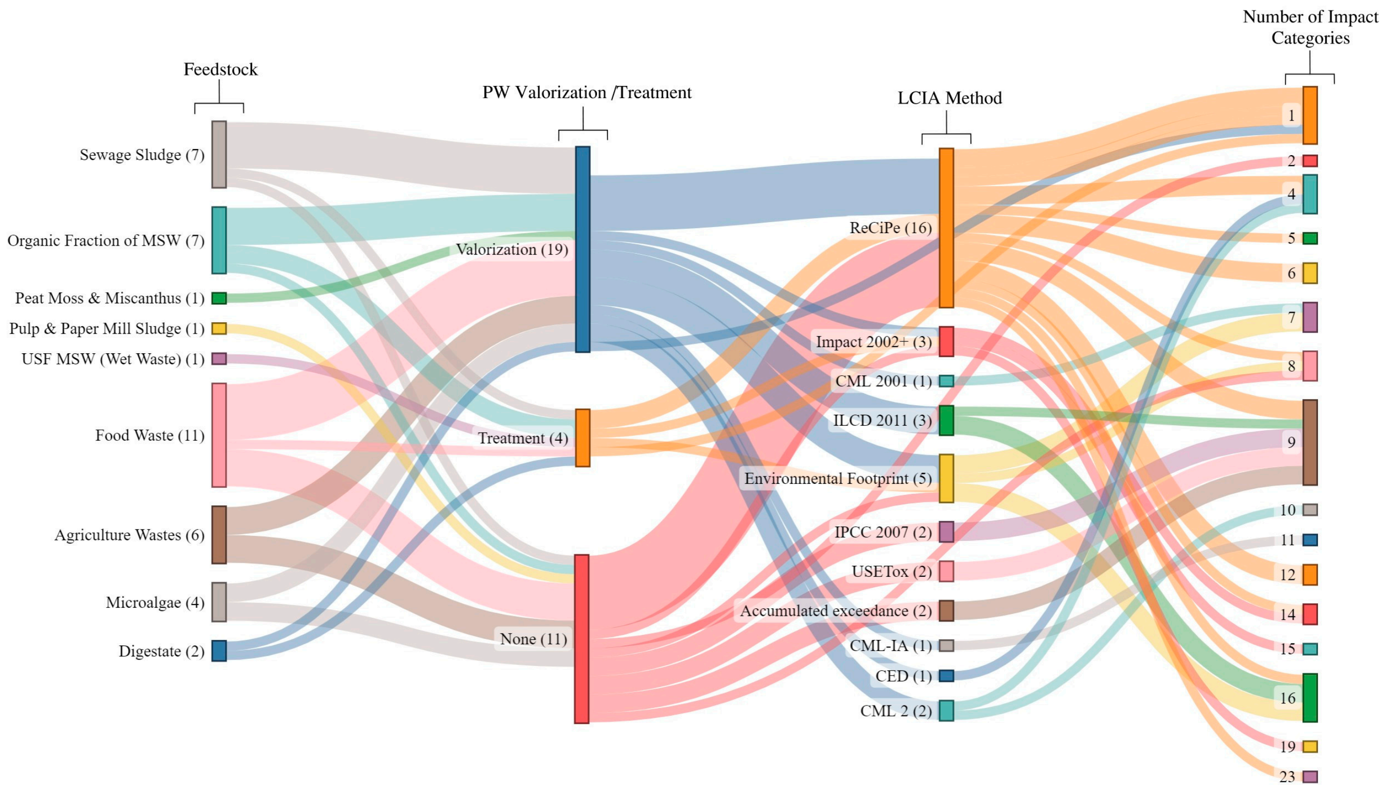

Figure 2. Sankey diagrams illustrating the connection between feedstock, valorization pathway, LCIA method, and number of impact categories for the collected studies for the process water. The numbers in parentheses represent the number of collected studies associated with each category in the figure.

Table 1. Summary of LCA-relevant details in the collected studies.

Feedstock	Study Goal [1]	Functional Unit	Ref.
Agricultural Waste Digestate	A life cycle assessment of this integrated process that recycles the agricultural waste digestate into H_2-rich syngas is carried out to evaluate the environmental impact	0.907 t of dry agricultural waste digestate and 1 MJ of syngas product	[26]
Almond Shells	Assess the technical and environmental performances of H_3PO_4-assisted HTC process chain existing at the laboratory scale, to eventually support its optimization and further upscale from an ecodesign perspective	0.4 g of activated carbon and 0.066 g of levulinic acid per gram of almond shell	[27]
Dairy Manure	Evaluate the environmental performance of novel nutrients, energy, and water innovations for resource recovery system intended to improve the sustainability of dairy manure management with an integrated HTC component	1000 kg wet manure produced on a largescale (1000 milking cows) CAFO	[28]
Dairy Manure	Compare the environmental impacts of resource recovery through NEWIR to existing manure management strategies and AD	1000 kg raw manure	[29]
Date Palm Fronds	Provide a comprehensive analysis of the environmental performance of the date palm HTC process	Use (i.e., processing) of 1 kg of palm waste biomass	[30]
Food Waste	Compare incineration, anaerobic digestion, and FWEB system in Singapore's context from an environmental perspective	1 t of food waste	[31]

Table 1. *Cont.*

Feedstock	Study Goal [1]	Functional Unit	Ref.
Food Waste	Conduct a system-level analysis to benchmark the environmental impacts of a combined HTC, nutrient recovery, and AD process for food waste valorization against the conventional stand-alone AD configuration	treatment of 1 kg of wet food waste	[24]
Food Waste	Evaluate the environmental impacts associated with the HTC of food wastes and the subsequent combustion of the generated solid product (HC) for energy production	1 kg of food waste	[32]
Food Waste	Compare hydrothermal carbonization with AD and composting to assess the energy and environmental advantages of the proposed system	1 t food waste	[33]
Food Waste and Organic Fraction of MSW	Analyze the environmental and economic performance of custom treatment paths (anaerobic digestion, HTC and incineration—in series and individually) for organic waste streams in Germany	1 kWh of energy	[34]
Green Waste	Provide a case study for the metropolitan region of Berlin, Germany in which leaves and grass cuttings are inputs for the HTC process	50,000 t/year of prepared and purified fresh matter	[17]
Green Waste, Food Waste, Organic Fraction of MSW, and Digestate	Present life cycle inventory and life cycle impact assessment results of HTC of green waste (being garden trimmings), food waste (represented by orange peels), organic fraction of municipal solid waste, and digestate at industry-relevant scales	1 MJ of heat to a building from a domestic 5–15 kW stove	[35]
Microalgae	Investigate the environmental performance and technical-economic viability of producing the standout products from microalgae—solid biofuel and biofertilizer—at a commercial level	12.5 m^3/h wastewater effluent	[36]
Microalgae	Perform environmental assessment to determine the environmental impacts of the production and utilization of HC blends and co-firing with hard coal.	1 kWh of generated electricity	[37]
Microalgae	Determine the environmental impact of the biological treatment of PW and nutrient recovery via struvite	1 m^3 of treated PW	[38]
Microalgae & Sludge	Assess the environmental performance of the different co-HTL processes, some of which involve the use of HTC	disposal of 100 kg of mixed materials on a dry basis, consisting of 50% sewage sludge and 50% microalgae	[39]
Olive Mill Waste	Determine the environmental impacts associated with TPOMW treatment using HTC, and to compare these impacts with those associated with currently used biological and thermal treatment approaches	treatment of 1 kg of fresh TPOMW	[40]
Olive Pomace	Transform hard-to-dispose-off olive pomace into HC via HTC and analyze the environmental impacts of post-process products	0.907 t olive pomace	[41]
Olive Pomace and Grape Marc	Identify which process, combustion, gasification, HTC, and pyrolysis is the most suitable alternative for grape marc and olive pomace	1 kg of pomace with 60% moisture	[42]
Organic fraction of MSW	Determine if HC is a more environmentally friendly energy carrier than lignite coal	production of 1 kWh of electricity	[43]
Organic Fraction of MSW and Sewage Sludge	Compare the environmental performance of three different blends of organic biomass wastes in the search for renewable and environmentally sustainable energies	1 kg of raw material	[44]

Table 1. *Cont.*

Feedstock	Study Goal [1]	Functional Unit	Ref.
Organic fraction of MSW & AD Digestate	Evaluate whether the HTC process can reduce the greenhouse gas emissions when treating OFMSW compared to the current base case	production and export of exergy	[45]
Organic Fraction of Urban MSW	Evaluate the environmental impact of extracting phosphorus from HC and using the HC as solid fuel	1 kg of wet biowaste with 100% content of biogenic carbon of total carbon	[46]
Peat Moss & Miscanthus	Evaluate the environmental performance of hydrothermally carbonized biomass (peat moss, miscanthus, and a blend of the two) used for energy or as a soil amendment compared with untreated biomass	1 t dry feedstock either left on-site or processed and used for either soil amendment or energy application	[19]
Poultry Litter	Determine the feasibility of using slow pyrolysis, fast pyrolysis, gasification, HTC, hydrothermal liquefaction, and supercritical water gasification specifically for the case of poultry litter and to determine whether they provide clear benefits over the conventional disposal method of direct land application	management of 1000 kg of fresh or wet poultry litter with a 25% w/w moisture content	[47]
Pulp & Paper Mill Sludge	Assess HTC and gasification as alternative treatments for the organic fraction of urban solid waste	(1) 0.907 t of the organic fraction of MSW and (2) 1 MWh energy	[18]
Rice Husk	Conduct a technical, economic, and environmental analysis of rice husk to fuel based on three conversion technologies: hydrothermal carbonization with palletization, catalytic pyrolysis, and anaerobic co-digestion	0.907 t rice husks	[48]
Sewage Sludge	Evaluate the environmental performance of an integrated system of an existing Water Resources Recovery Facility and a hypothetical hydrothermal carbonization plant applied to the generated sewage sludge	1 t of sewage sludge	[49]
Sewage Sludge	Evaluate the feasibility and efficiency of integrating an HTC section into the layout of a conventional WWTP	1 m^3 of wastewater entering the plant	[50]
Sewage Sludge	Compare the environmental performance of the HTC and AD-integrated scenario with the standalone AD system for sewage sludge valorization	treatment and disposal of 1000 kg wet mixed sludge consisting of 60.4% PSS and 39.6% SSS	[4]
Sewage Sludge	Provide scientifically substantiated evidence of whether pre-treatment by HTC or pyrolysis can give the decisive edge in reducing environmental impacts in comparison to a direct combustion of sewage sludge	1 t of raw sewage sludge	[51]
Sewage Sludge	Evaluate the environmental consequences of different alternatives for using HC pellets produced from mixed sludges from pulp and paper mills in Sweden	1 t of dry sludge	[10]
Sugracane Bagasse	Quantify environmental impacts of electricity production from sugarcane bagasse HC generated via microwave-assisted HTC	1 MJ electricity generation HC produced from MAHTC treatment	[52]
Wet Mechanically Separated MSW	Clarify the sustainability of the HTC process at a system level from an environmental point of view	1 t of USF	[53]

[1] The study goal listed in this table is summarized and abbreviated from that found in each article.

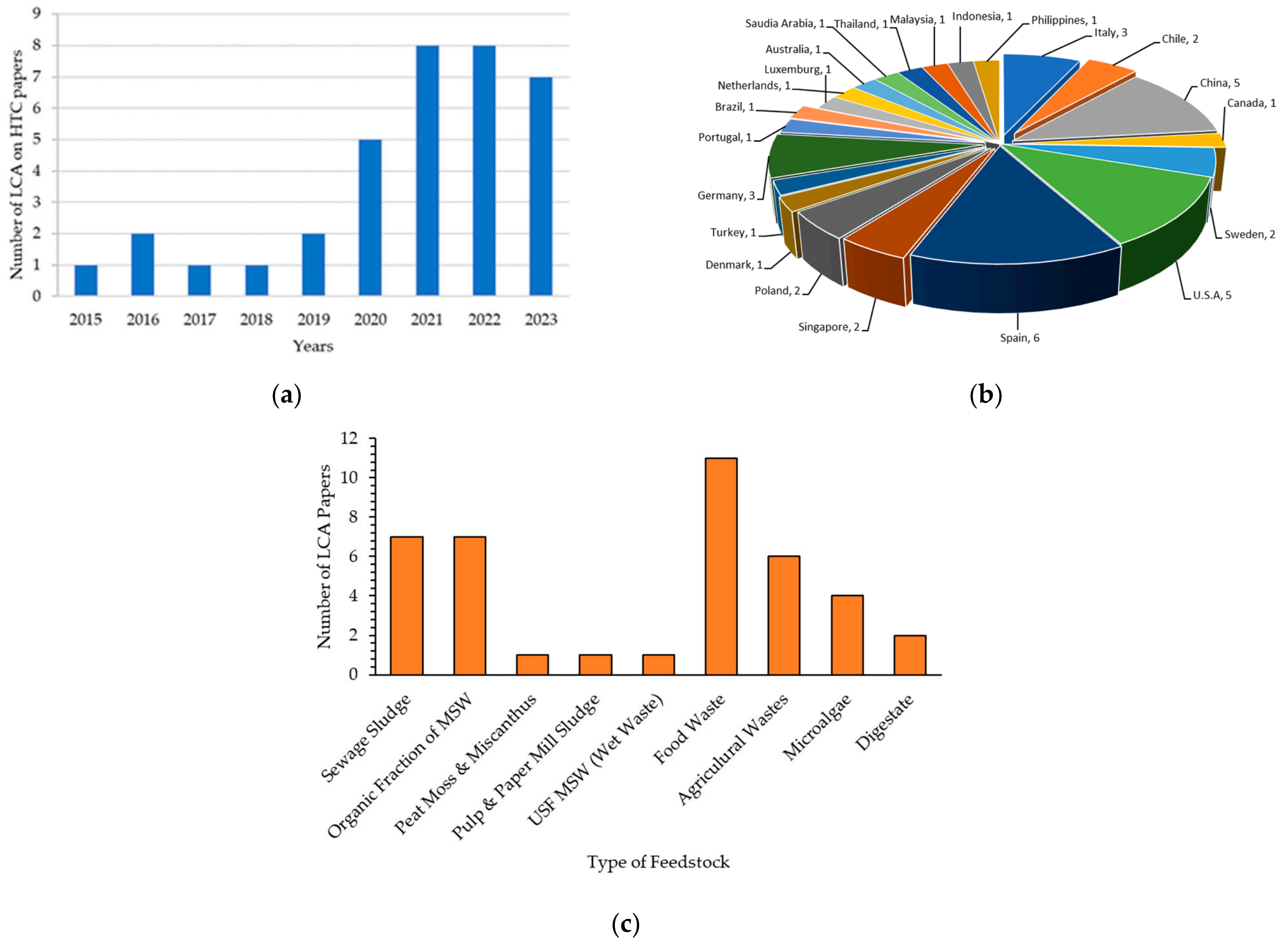

Figure 3. Data illustrating trends associated with the collected LCA studies: (**a**) Number of LCA publications over time; (**b**) Geographical regions in which the LCA studies originated (numbers represent the number of collected studies); (**c**) Feedstocks associated with the different LCA papers. USF = undersieved fraction.

Overall, all LCA papers collected in this effort, with the exception of one, included some assessment of the environmental implications associated with product valorization (either HC or PW). HC valorization was conducted in 91% of the collected studies. Figure 4 shows the specific HC valorization pathways explored in these studies, illustrating that the majority involved HC conversion to energy. PW valorization has been less studied, with 59% of the collected studies including this aspect. It should be noted that, if not valorized, PW treatment was included in the majority of studies; only 35% of the studies did not account for the treatment/valorization of PW. Although possible, none of the studies included gas valorization. Sztancs et al. [37] suggested valorization of the gas would be completed in their integrated system, but the valorization process remained outside the boundaries of their LCA study. In other studies, the gas stream was either released to the atmosphere or treated prior to discharge.

Specific details associated with the LCAs conducted in these studies are also included in Table 1 and Figures 1 and 2. The majority of these studies focused on comparing HTC with other, more traditional waste management processes (e.g., incineration, composting). A few exceptions to this existed. One exception was a study by Chaparro-Garnica et al. [27], whose study used the HC as an environmental adsorbent. The impact assessment conducted in these studies generally included four or more impact categories (Figures 1 and 2), with 47% of the papers utilizing some version of the ReCiPe impact assessment method.

Only four of the collected studies investigated a single impact category (global warming potential). The functional units used in these studies were almost all based on the treatment and/or management of some mass/volume of a waste stream; eight of the collected studies used functional units associated with energy generation and one was associated with activated carbon production.

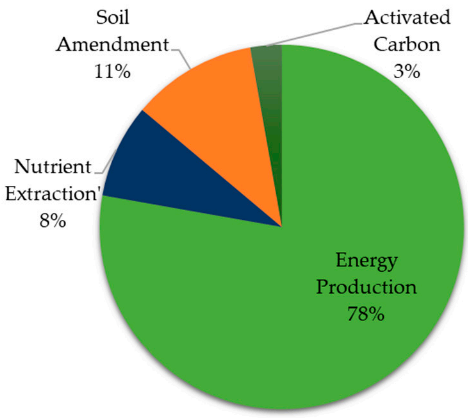

Figure 4. HC valorization pathways associated with the LCA studies collected in this effort.

3.2. Review of HTC Product Inclusion in Collected LCA Studies

The goal of this component of the review is to gain an understanding of how product valorization is currently being modeled in LCA studies. Specific details associated with the LCAs surrounding the HTC process and/or results from comparisons between HTC and other processes were not reviewed in this effort. Recent review papers on these topics provide additional information about these aspects of LCA studies [20–23].

3.2.1. Hydrochar Valorization

HC conversion to energy represents the valorization pathway most included in HTC-related LCA investigations. The overwhelming interest in this valorization pathway is not surprising, as the majority of experimental studies in this area have focused on energy-related implications [13,54,55]. Overall, results from these LCA studies indicate that HC combustion has the potential to result in significant environmental benefits. Mannarino et al. [49] and Mohammadi et al. [10] reported that substituting lignite-derived and coal-derived energy, respectively, with HC-derived energy resulted in significant avoided environmental impacts.

Table 2 contains a summary of the collected studies that included HC conversion to energy, specifically highlighting the series of processes required to achieve energy generation included in the models and, if applicable, the fuel source substituted by the HC-based energy. In the vast majority of these studies, energy generation was achieved via HC combustion/incineration. One exception to this was the study conducted by Corvalán et al. [18], in which energy was generated via HC gasification. A few major differences between these studies exist. One difference was associated with the approaches used to prepare the HC for combustion/incineration, which ranged from no HC preparation to a multi-step preparation process including dewatering, solid/liquid separation, drying, pelletization and transport. Of these processes, the majority of the collected studies included some sort of separation of the HC and PW, followed by HC drying. Data associated with these processes ranged from primary experimentally derived data (e.g., HC yields, filtration efficiencies) to secondary data based on study-dependent assumptions (e.g., pelletization). Another difference between these studies was how emissions from HC combustion were modeled. Combustion emissions were often assumed to equal emissions from the combustion of other waste streams, such as municipal solid waste, sludge, or coal [10,32]. Exceptions to this included the study conducted by Owsianjak et al. [35], Sarrion et al. [24], and Oliver-Tomas et al. [46], who all used emissions from a pilot-scale plant. The fuel sources substituted by the HC-based energy, summarized in Table 2, also differed between studies, and ranged from biomass to lignite coal. Berge et al. [32], Zhang et al. [52], and Mohammadi et al. [10] reported the importance in the source of fuel being substituted and the impact such choices can make in the overall conclusions, indicating the importance of modeling conditions specific to each scenario. Substituting a green energy source, such as biomass, is likely to have a smaller positive environmental impact (or possibly be deemed disadvantageous) than when substituting a coal-based energy source. Sztancs et al. [37] investigated the environmental implications associated with co-firing HC with hard coal, and found that greater HC fractions in the HC/coal blend were environmentally advantageous.

Table 2. Summary of HC to energy processes modeled in the collected studies.

Feedstock	HC Processing Steps Included	Fuel Source Substituted	Ref.
Dairy Manure	Drying, pelletizing, combustion	Low-grade coal in conventional coal-fired power plants	[28]
Dairy Manure	Filtration, drying, combustion	coal	[29]
Food Waste	Drying and combustion	hard coal briquettes	[24]
Food Waste	Solid/liquid separation, drying, pelletization, combustion	lignite	[34]
Food Waste	Drying, combustion	Compared different coals, biomass, and average US electricity mix	[32]
Food Waste and Organic Fraction of MSW	Drying	hard coal briquettes	[4]
Green Waste	Distribution, combustion	Hard coal in electricity and heat	[17]
Microalgae	HC separation	No substitution appears to have been modeled	[36]
Microalgae	HC dewatering (centrifugation and heating), combustion in a combined heat and power plant	Hard coal, as a result of blending with HC	[37]
Olive Pomace	Centrifugation, drying, combustion	Electricity	[41]
Olive Mill Waste	Gravity drainage, drying, incineration	Coal-based electricity	[40]
Olive Pomace and Grape Marc	Dryer, gasifier	No substitution appears to have been modeled	[42]
Organic Fraction of Urban MSW	De-ashing, drying, pelletizing, combustion	fossil coal	[49]
Organic fraction of MSW	Decanter, belt dryer, pelletizer, incineration in lignite power plant	Lignite coal	[43]
Organic Fraction of MSW and Sewage Sludge	Filtration, drying, pelletization, combustion	Chilean energy matrix	[44]
Organic Fraction of MSW and AD Digestate	Solid/liquid separation, drying, pelletizing, incineration or gasification	No substitution appears to have been modeled, HC was co-combusted with lignite coal	[45]
Peat Moss & Miscanthus	Drying and transportation	No substitution appears to have been modeled	[19]
Pulp & Paper Mill Sludge	Filtration, drying, pelletization, combustion	No substitution appears to have been modeled	[18]
Rice Husk	Filtration, drying, pelletizing	Not reported	[48]
Sewage Sludge	Drying (belt press and heat), pelletization, combustion	Lignite	[49]

Table 2. *Cont.*

Feedstock	HC Processing Steps Included	Fuel Source Substituted	Ref.
Sewage Sludge	Mechanical dewatering by filter press, drying, incineration	No substitution appears to have been modeled	[50]
Sewage Sludge	Dewatering, drying, pelletization, combustion	Compared substitution of coal and solid wastes	[10]
Sewage Sludge	Centrifugation, drying, pelletizing, combustion	fossil fuel	[53]
Sewage Sludge	Dewatering, transport, incineration	German electricity mix	[51]
Sugracane Bagasse	Briquette production, transport, emissions from combustion	Compared several: average high voltage electricity generation, electricity from co-generation of sweet sorghum bagasse, incineration of MSW, conventional natural gas power plant, hard coal combustion, and lignite combustion	[52]
Wet Mechanically Separated MSW	De-ashing, drying, pelletizing, combustion	Fossil coal	[35]

Other approaches modeled to gain energy from HC included those in which HTC was integrated with other processes, such as hydrothermal liquefaction (HTL) and pyrolysis. Wen et al. [26] modeled the utilization of HTC as a pretreatment process in which HC was generated and subsequently pyrolyzed to a hydrogen-rich syngas (it was assumed the hydrogen would be used in energy applications). In addition, Wen et al. [26] included the use of pyrolyzed HC as a sink for carbon. Zhang et al. [39] investigated the valorization of HC by using it in HTL processes to ultimately generate a bio-oil.

The approaches used in these studies to model energy generation from HC highlight some important weaknesses that require further attention/study. In each study, the specific amount of energy generated was determined based on either the HHV or LHV of the HC (along with HC yields) and assumed generation was 100% efficient. It is unlikely that this assumption is valid during actual HC combustion. For example, Ischia et al. [56] and Lucian et al. [57] report the possible need for secondary char removal for adequate HC combustion, which may impact both the pretreatment steps required for HC combustion, as well as the overall amount of energy that can be derived from the process. Inclusion of such details is important to consider in future LCA investigations to ensure more realistic scenarios are modeled. Additionally, more work understanding the environmental aspects associated with HC preparation prior to combustion is important to understand. For example, understanding if important differences between mechanical dewatering vs. gravity filtration exist is critical in developing guidance on process optimization.

Another valorization pathway explored in these collected papers is the use of HC in agricultural applications, including nutrient extraction from the HC or direct use of the HC as a soil amendment. This pathway, from an LCA perspective, appears to still be in its infancy. Few of the collected LCA studies included this valorization approach. Nutrient extraction from the HC was modeled via two approaches. One approach involved phosphorus extraction via acid washing [46,49], usually after the HC was separated from the PW. Nutrient extraction has also been proposed from the ash resulting from HC combustion [51]. Table 3 summarizes the studies in which this approach was modeled and includes the processes associated with the acid washing and the nutrient source the HC-derived nutrients replaced. Acid addition is required in this approach. Results from LCA studies indicate that this may be problematic from an environmental perspective. Mannarino et al. [49] found that the use of nitric acid for phosphorus leaching resulted in large overall environmental impacts in comparison to relatively small contributions from any avoided impact resulting from the recovered phosphorus. Oliver-Tomas et al. [46]

investigated the use of a range of acids for phosphorus leaching, but reported that the use of HC for both energy generation and phosphorus recovery represented an overall environmental burden.

Table 3. Summary of the nutrient extraction processes modeled in the collected studies.

Feedstock	Processes Included in the Extraction	Extraction Acid	Nutrient Replacement Sources	Ref.
Organic Fraction of Urban MSW	Drying, acid and base addition, land application	Compared HNO_3, HCl, H_2SO_4	Phosphorus from phosphate rock	[46]
Sewage Sludge	Filter press, dryer, grinder, mixer (HNO_3), filter press, land application	HNO_3	Phosphorus fertilizer	[49]
Sewage Sludge	Drying, HC incineration, phosphorus recovery from ash	Not reported	Not reported	[51]

Using the HC as a soil amendment has garnered significant interest in the HTC community, resulting in many lab and field explorations. However, only 4 of the collected LCA studies included the use of HC as a soil amendment (Table 4). In all studies, it was assumed that after HC separation from the PW and/or drying, it would be directly land applied and would substitute (at a ratio of 1:1) chemically based nitrogen fertilizers, which is unlikely. Some type of HC preparation is likely necessary to remove toxic compounds found on the HC surface [56,58,59]. Roy et al. [19] and Bora et al. [47] included an estimate of carbon mineralization following land application in their LCA models. Understanding carbon dynamics is critical in understanding the application of appropriate credits/offsets. Roy et al. [19] assumed a typical first-order carbon mineralization rate to estimate carbon sequestration. In both cases, significant assumptions were made in this aspect of the model; data to validate these assumptions are currently lacking. Obtaining such data is critical in assessing potential long-term environmental benefits associated with using the HC as a soil amendment and represents a current research gap.

Table 4. Summary of the collected studies in which HC was used as a soil amendment.

Feedstock	Processes Modeled	Ref.
Food Waste	Filtration and drying	[33]
Microalgae	Filtration	[36]
Peat Moss and Miscanthus	Transport, land application with carbon mineralization	[19]
Poultry Litter	Land application (including mineralization of carbon)	[47]

Overall, these studies indicate that there are potential environmental benefits associated with using HC in agricultural applications, but they are dependent on several assumptions and site-specific details. Roy et al. [19] reported that HC used as a soil amendment was more environmentally beneficial than using it as an energy source, but this conclusion was significantly dependent on the assumed decomposition rate of biomass. Similarly, Zhao et al. [33] indicated that HC-derived soil amendments have superior carbon fixation compared to land applied compost and digestate. Castro et al. [36] reported that using HC as a soil amendment required an energy input 2.5 times less than that associated with using it as an energy source and, correspondingly, reported that using HC as a soil amendment resulted in a lower or equal ecological footprint than HC-derived energy across all impact categories analyzed.

Other applications for the HC have been proposed in the literature, such as using the HC as an adsorbent. Chaparro-Garnica et al. [27] conducted an LCA study investigating the environmental impacts of converting HC generated from the carbonization of almond shells to an activated carbon via H_3PO_4 activation (activation occurred within the system). Results indicated that electricity needs and pyridine, needed for functionalization, were the processes responsible for the greatest impacts on the environment. It should be noted that Chaparro-Garnica et al. [27] used LCA results to redesign their process and reduce environmental impact. Involving LCA in technology development has been shown to result in more environmentally beneficial processes [60,61].

3.2.2. Process Water Valorization

Almost all the collected LCA studies included PW treatment and/or valorization (Table 5). Only 35% of the collected studies assumed no treatment and that the PW was directly discharged to the environment. Although these specific scenarios were not realistic, they did provide information associated with the environmental impact associated with the PW. A significant fraction of the studies (Table 5) assumed the PW would be treated either at an existing wastewater treatment plant (WWTP) or through more complex processes such as reverse osmosis and/or nanofiltration. In this review, it was assumed that any PW treated at a WWTP did not involve valorization. Reverse osmosis and/or nanofiltration treatment processes were counted as valorization pathways if the resulting materials (e.g., concentrate) were collected and then used in some beneficial way, such as a soil amendment [35] or energy source [34,51]. Owsianjak et al. [35] reported that using the PW as a fertilizer did reduce impacts from virgin fertilizer production, but the extent of the reduction was small; the amount of fertilizer gained from the PW was small in comparison to that needed for their modeled application, thus the amount of conventionally produced fertilizer remained significant.

Table 5. Summary of PW treatment and/or valorization in the collected studies.

Feedstock	PW Management	Ref.
Agricultural Waste Digestate	No treatment	[26]
Almond Shells	Extraction of levulinic acid using rotary evaporation, following extraction the liquid is used as input to solid-phase activation	[27]
Dairy Manure	Algae cultivation for nutrient recovery and production of protein-rich cattle feed, and water recovery from algae pond effluent via membrane distillation	[28]
Dairy Manure	Algae cultivation for nutrient recovery and production of protein-rich cattle feed, and water recovery from algae pond effluent via membrane distillation	[29]
Date Palm Fronds	No treatment	[30]
Food Waste	Transesterfication of bio-oil by acid treatment and glycerol recovery	[31]
Food Waste	Nutrient recovery via struvite, then AD with biogas generation and combustion and subsequent land application of digestate	[24]
Food Waste	No treatment	[32]
Food Waste	Treatment at a WWTP	[33]
Food Waste and Organic Fraction of MSW	Nanofiltration/reverse osmosis and combustion of the retentate	[34]
Green Waste	Treatment at a WWTP	[17]
Green Waste, Food Waste, Organic Fraction of MSW, and Digestate	Reverse osmosis; concentrate is diluted and then land applied	[35]
Microalgae	No treatment	[36]

Table 5. *Cont.*

Feedstock	PW Management	Ref.
Microalgae	No treatment	[37]
Microalgae	Biological treatment followed by struvite precipitation; precipitates to be used as a fertilizer	[38]
Microalgae & Sludge	Treat via HTL to produce products of value	[39]
Olive Mill Waste	No treatment	[40]
Olive Pomace	AD with biogas generation and combustion	[41]
Olive Pomace and Grape Marc	No treatment	[42]
Organic fraction of municipal solid waste	Nanofiltration and reverse osmosis; excess permeate was sent to a WWTP and excess retentate was combusted	[43]
Organic Fraction of Urban MSW	Reverse osmosis; concentrate is diluted and then land applied	[46]
Organic Fraction of MSW and AD Digestate	Nanofiltration and reverse osmosis; excess permeate was sent to a WWTP and excess retentate was combusted	[45]
Organic Fraction of MSW and Sewage Sludge	No treatment	[44]
Peat Moss & Miscanthus	Direct land application	[19]
Poultry Litter	AD with biogas generation and combustion and subsequent land application of digestate	[47]
Pulp & Paper Mill Sludge	No treatment	[18]
Rice Husk	No treatment	[48]
Sewage Sludge	AD with biogas generation and combustion and subsequent land application of digestate (following composting)	[49]
Sewage Sludge	AD with biogas generation and combustion	[50]
Sewage Sludge	Treatment at a WWTP	[10]
Sewage Sludge	AD with biogas generation and combustion and subsequent land application of digestate	[4]
Sewage Sludge	Nanofiltration and reverse osmosis; excess permeate was sent to a WWTP and excess retentate was combusted	[51]
Sugracane Bagasse	Not included in the analysis	[52]
Wet Mechanically Separated MSW	Comparison between WWTP, reverse osmosis, AD and a combination of AD and reverse osmosis	[53]

AD, followed with biogas collection and/or land application of the digestate, was a common pathway for PW valorization. Mannarino et al. [49] and Yay et al. [41] reported the positive benefits associated with the AD of the PW. Yay et al. [41] compared PW treatment at a WWTP with PW valorization via AD, and reported on the benefits associated with including the AD process. Lombardi et al. [53] compared AD, reverse osmosis, and a combination of AD and reverse osmosis for PW valorization, and reported that AD generally improved system environmental impact because of biogas recovery, while the inclusion of reverse osmosis added additional process requirements that resulted in a greater environmental burden. Importantly, Lombardi et al. [53] concluded that the burdens associated with PW treatment are small in comparison with the overall benefits associated with the HTC process.

Other strategies for PW valorization have been studied. Glover et al. [28,29] incorporated HTC and PW valorization into the management of dairy farm waste. In each study, the PW was used for algae cultivation for nutrient recovery and production of protein-rich cattle feed, and water recovery from algae pond effluent via membrane distillation. Each study details the positive impact this valorization imparted to their integrated system. In another study, acid-mediated HTC was conducted, with the goal of enhancing nutrient extraction during the HTC process [24]; a nutrient-rich PW resulted. Sarrion et al. [24] recovered phosphorus from the PW via struvite precipitation, which could then be land applied. Land application of this material was modeled to replace a conventional fertilizer, which resulted in net environmental benefits to the system. Roy et al. [19] modeled an approach that involved the direct application of the PW on land as an irrigation source/fertilizer. Ahamed et al. [31] modeled an approach in which the PW was upgraded to biodiesel, which was shown to be an advantageous valorization route of food waste. Chaparro-Garnica et al. [27] investigated the environmental impacts associated with recovery of levulinic acid from the PW and found the environmental impacts from this to be small, suggesting that chemical recovery from PW may be environmentally advantageous. Zhang et al. [39] investigated the valorization of PW by using it in HTL processes to ultimately generate a series of value-added products. Hu et al. [38] focused their LCA study on PW treatment; they investigated its biological treatment followed by struvite precipitation and found that using the precipitated nutrients was environmentally beneficial when offsetting the need for fertilizer.

3.3. Research Gaps and Needs

The majority of the LCA studies reviewed investigated the environmental impacts of the HTC process in comparison with the environmental impact of another waste management process. Results generally indicated that HTC was more environmentally beneficial than the traditional processes in which comparisons were being made, in large part because of the value gained from product use (and thus avoided virgin product needs). These collected LCA studies highlight the importance of HTC product valorization and some important environmental aspects associated with the investigated valorization processes. They also indicate the need for additional work in this area. Conducting LCA studies that probe the important factors and conditions controlling the environmental impact of product valorization is critical and would provide information necessary for the generation of specific HTC-related policies. When considering HC valorization, this involves gaining a deeper understanding of the following:

- Full system level integration to evaluate the influence of location-specific information (e.g., local energy infrastructure, transport distances) on HC valorization impacts;
- HC pre-processing needs for efficient energy generation and land application, and the impact those processes may have on the environment;
- Guidance associated with minimizing environmental impact during HC processing/preparation for energy generation and land application;
- Delineating how critical HC properties (e.g., conversion efficiency to energy) influence product valorization. These are critical in the development of guidelines to achieve environmentally beneficial systems;
- HC behavior in the environment after land application (e.g., impacts on crop growth, carbon sequestration).

There are also needs associated with understanding PW valorization, which involves gaining a deeper understanding of the following:

- Full system level integration to evaluate the influence of critical factors that control environmentally-beneficial PW valorization approaches;
- Influence of location-specific information (e.g., local infrastructure) on PW valorization;
- Identify the PW properties most critical when deciding between the valorization approaches;
- PW pre-processing (e.g., dilution) requirements for different valorization pathways.

In the current body of LCA studies, no gas valorization was conducted. Thus, there is a need for conducting some studies to probe what valorization processes are viable (e.g., energy generation, chemical recovery) and whether the approaches are environmentally beneficial.

It should also be noted that there is a distinct need for more data related to product valorization. The majority of LCA studies reviewed in this effort used some laboratory-derived primary data coupled with significant secondary data. Secondary data was mostly associated with HTC product application in the environment. Primary data associated with these processes are needed. In addition, data from larger-scale HTC efforts are needed. More collaborations with the HTC companies currently operating at larger scales would be extremely beneficial in providing more validated operational guidance to minimize environmental impacts.

4. Conclusions

Studies describing HTC-related LCA efforts were collected with the goal of gaining an understanding of how HTC product valorization is currently being modeled in LCA studies and to use that information to assess current research and/or data needs associated with product valorization. Results indicated that HC conversion to energy was the most modeled valorization pathway. Approaches in which the energy was generated differed between studies. Other HC valorization approaches included nutrient extraction, use as a soil amendment, and generation of an activated carbon. Not all studies included PW valorization; most included valorization or some sort of treatment. PW valorization pathways differed significantly between studies, with the majority converting PW organics to biogas via AD. From this review of the literature, it is clear that the potential exists for HTC product valorization. To realize this potential in a full-scale application, research gaps and data needs were identified that included a system level integration to evaluate location-specific information as well as more extensive characterization of the impact of HTC product properties on valorization impacts.

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