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Life Cycle Assessment and Technoeconomic Analysis of Thermochemical Conversion Technologies Applied to Poultry Litter with Energy and Nutrient Recovery

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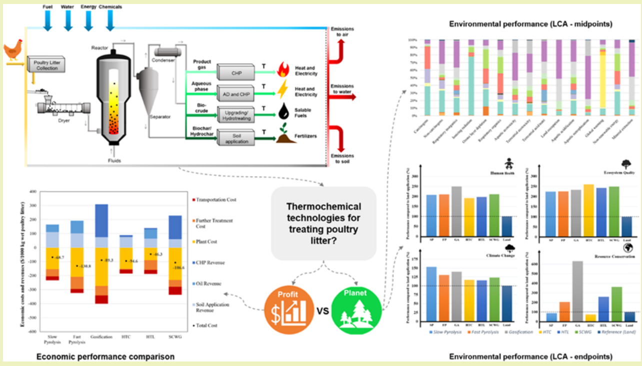

ABSTRACT: Thermochemical technologies provide promising pathways to recover energy and reduce environmental impacts from biomass wastes. Poultry manure or litter additionally provides an opportunity for recovering and recycling nutrients and producing valuable soil amendments. This study compared the life cycle environmental impacts and technoeconomic performance of six thermochemical technologies for treating poultry litter waste—slow pyrolysis, fast pyrolysis, gasification, hydrothermal liquefaction, hydrothermal carbonization, and supercritical water gasification—with direct land application. Using life cycle assessment (LCA), the technologies were compared through 15 different environmental impact categories (midpoints) using the IMPACT 2002+ method. On converting the midpoints to damage categories (end points), it was found that these technologies outperformed the conventional land application method with respect to human health (92−149% improvement), climate change impact (15−53% improvement), ecosystem quality (124−160% improvement), and resource depletion (−24−530% improvement). The technoeconomic analysis (TEA) identified carbon price (breakeven of $127/1000 kg CO_2 equiv for slow pyrolysis) and high capital costs as influential parameters for large-scale applications of these technologies. The TEA results were most sensitive to carbon price and transportation distance (0.69 and 0.52% changes in revenue per change in input, respectively).

KEYWORDS: *life cycle assessment, technoeconomic analysis, thermochemical technologies, poultry litter, waste-to-energy*

■ INTRODUCTION

According to the recent Paris Agreement and IPCC reports, all possible pathways to limit temperature rise to 1.5 °C involve the utilization of considerable renewable energy resources along with some form of carbon capture.[1] Bioenergy is a key component in most of these mitigation pathways.[2,3] However, there have been frequent debates about the sustainability of energy crops for biofuel production, in light of their associated land use impacts and the competition with traditional crops for water and energy.[4−6] Waste-to-energy processes, on the contrary, provide a unique opportunity for simultaneously producing energy and disposing of wastes,[7] but many of them have not been sufficiently investigated.[8]

Processes such as incineration and anaerobic digestion have been popular in this field, but there have been questions raised about their impact on the environment, with concerns regarding air pollutants released through the former and the impact on water bodies through the digestate produced from the latter.[9,10] Emerging thermochemical technologies such as gasification (GA), pyrolysis, and hydrothermal processes, on the other hand, may have the potential to treat wastes with minimum environmental impact and produce useful products in different forms, with applications in both energy generation and nutrient recycling.[11,12] However, limited information exists as to how their environmental performance or energy balance is compared to that of direct land application or with each other.

Poultry litter, generated through intensive poultry production, is one of the most abundant animal wastes globally because of the increasing demand for poultry meat and egg products (with the global poultry population estimated at nearly 22 billion in 2010).[13] The high nitrogen (N), phosphorous (P), and potassium (K) contents make poultry litter a desirable fertilizer as these nutrients are energy-intensive to produce otherwise and consume considerable resources.[14−16] However, the traditional disposal pathway of

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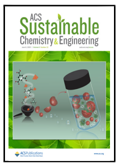

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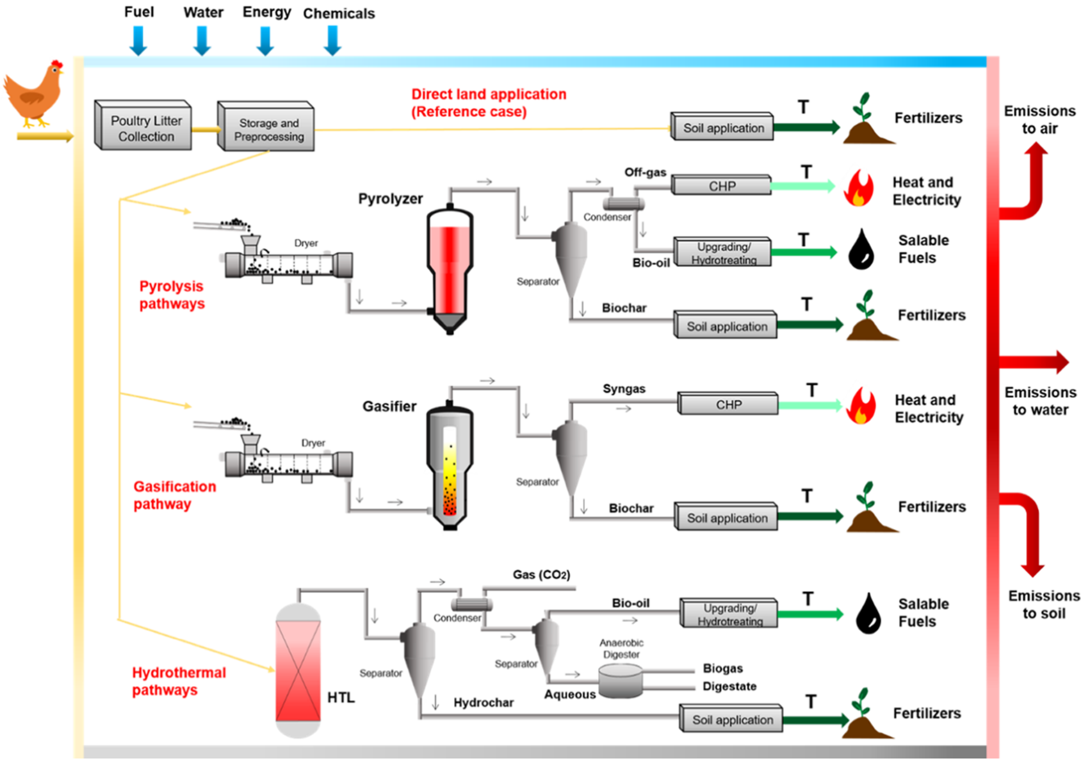


Figure 1. System boundaries and components for the different scenarios considered through a schematic representation. The reactors, product separation, and product distribution for each process were different. The digestate from the AD was further utilized as a fertilizer, and this is not shown in the figure (T = transportation, AD = anaerobic digester, CHP = combined heat and power generation).

direct land application of poultry litter has a large environmental footprint due to issues such as eutrophication, spreading of pathogens, antibiotic residue accumulation, and greenhouse gas (GHG) emissions among others.[17,18] Thus, there is an urgent need for developing scalable methods to treat organic wastes such as poultry litter while considering resource recovery if sustainability goals are to be met.[19−23]

There have been some studies based on the comparative environmental evaluation of different treatment methods for pig manure and dairy manure in which certain biological and thermal methods were compared with those of direct land application.[24] For poultry litter, although the experimental information for thermochemical technologies is still incipient, previous pilot-scale operations have revealed the feasibility and potential environmental benefits through their utilization. Certain studies have also evaluated the environmental impacts of technologies such as gasification with the direct land application of poultry litter.[25] However, there is a need to identify how multiple conversion technologies and process schemes for such high-nutrient-containing wastes may be optimized to minimize all environmental burdens, including but also beyond climate change. Therefore, the primary objective of this work was to determine the feasibility of using certain thermochemical technologies (slow pyrolysis (SP), fast pyrolysis (FP), gasification, hydrothermal carbonization (HTC), hydrothermal liquefaction (HTL), and supercritical water gasification (SCWG)), specifically for the case of poultry litter and to determine whether they provide clear benefits over the conventional disposal method of direct land application. This in turn required special attention to the flow of products and nutrients through the system as they could be extremely important for the comparison. These particular technologies were chosen, as they are currently the most promising thermochemical technologies for treating poultry litter.[26−28]

Key novelties of this work included:

- The consideration of the six most promising thermochemical technologies whose performances have, to the best of our knowledge, not been compared in a single study for a common case such as poultry litter valorization and with respect to direct land application.
- The emphasis on determining the optimum pathways for the solid products obtained from each of the technologies using high-nutrient-containing waste feedstock.
- The detailed analysis of the nitrogen flow and related impacts, including but not limited to climate change within the systems on the environmental and economic performance of the technologies.

■ MATERIALS AND METHODS

Thermochemical Technologies Analyzed. The selection of the technologies for this analysis was based on a rigorous literature review, and Section S1 in the Supporting Information (SI) provides the relevant details along with a table summarizing the processes. Gasification is carried out at elevated temperatures in a restricted oxygen environment, and it is acknowledged for being cleaner than direct combustion due to syngas production.[29] Pyrolysis has also garnered much attention in the waste-to-energy field, mainly due to one of its solid products termed “biochar”, which possesses the potential for long-term carbon sequestration.[30−32] Pyrolysis can further be split into slow pyrolysis (reaction temperature of 400−600 °C with retention time in minutes to hours) and fast pyrolysis (reaction temperature of 500 °C or lesser and a shorter retention time of milliseconds to seconds).[33] Slow pyrolysis preferred when producing biochar is the primary goal, whereas biocrude is the primary product in fast pyrolysis. Hydrothermal processes are relatively new technologies to convert wet biomass into biofuels using water in a subcritical or supercritical state. These technologies are known to eliminate the energy-intensive step of predrying.[34−36] For technologies such as pyrolysis and gasification, which require 10−15% moisture content (wt %) to function optimally, the drying stage is essential unlike hydrothermal technologies. However, since poultry litter is a comparatively dry organic waste (20−25% moisture content on a wet weight basis),[26] the drying energy load is not as much as other organic waste streams.[37]

Collection of Data and Assumptions. *Thermochemical Technologies for Poultry Litter Valorization.* To maximize the recovery of energy and nutrients, we made the following assumptions (Table S1 in the Supporting Information). We considered that the solids produced from pyrolysis, gasification, and hydrothermal processes, and the digestate from anaerobic digesters (ADs) were applied to the soil. Biocrude produced from the technologies proceeded to upgrade facilities to produce diesel and gasoline. While the nitrogen concentration within the bio-oils from poultry litter is high, this is not necessarily true regarding the sulfur contents (less than 1%). There have been multiple studies supporting the feasibility of upgrading bio-oil produced from organic feedstocks very similar to that of poultry litter.[38,39] Product-gas (if produced in large volumes) was sent to a combined heat- and power-generating (CHP) station. The aqueous phase products, mainly produced from hydrothermal processes, were sent to AD−CHP units to recover energy and nutrients. It was assumed that the aqueous phase from the hydrothermal processes is sufficiently dilute to be directly fed to the ADs. Only partial recovery of nutrients would be possible through the AD as it has been found that around 30% of the organics from the HTL aqueous phase cannot be recovered through conventional anaerobic digestion.[40] Additionally, the ADs have the disadvantage of long residence times and low operating temperatures (which cannot ensure that all of the pathogens in the waste have been removed).[41] Hence, in this paper, the direct use of AD to treat poultry litter was not pursued and it was only coupled with the hydrothermal technologies through the aqueous phase.[36] The gases from both the pyrolysis technologies were combusted on-site with selective catalytic reduction (SCR) to remove NO_x at a 90% removal efficiency, and gas leaks were not considered in this analysis and could be a future addition to this work. The environmental performance of the conventional treatment approach for poultry litter, which employs direct application on land, was also evaluated (Figure 1) to provide a reference case as it is currently the most popular disposal method for poultry litter. Other developed technologies such as anaerobic digestion and incineration were excluded from the analysis as previous studies comparing these technologies have highlighted their limitations for treating poultry litter in terms of low yields and poor economic performance, as well as encouraged future work on exploring alternatives such as thermochemical technologies.[42]

To improve the quality of this assessment, mass and energy balances were utilized for the major processes in the system, and the values obtained were used in combination with data from the literature, subject to their availability. The carbon (C) and nitrogen (N) balances for the processes were incorporated into the calculations too (Figures S1 and S2, and Table S2 in the Supporting Information). In this study, 1000 kg of wet poultry litter was assumed to contain on average 14 GJ primary energy, 350 kg C, 46 kg N, 12 kg P, and 20 kg K.[26,43] The moisture content was assumed to be 25%[26,37] on a wet weight basis (additional information in the Supporting Information).

Soil Products. All of the thermochemical technologies produce characteristic solid products that could potentially be utilized as soil amendments (Table S4). In the case of direct land application of poultry litter, only collection and slurry-spreading activities were considered, and the environmental burdens were composed of greenhouse gas (GHG) emissions and leaching.[44−49] For the thermochemical technologies, although the properties of biochar vary based on the operating conditions and feedstocks, its effectiveness as a soil fertility enhancer has been extensively investigated, including for the biochar produced using poultry litter as input. In our work, the poultry litter biochar was assumed to have an H/Corg ratio of 0.358.[50] This corresponded to a mean residence time of almost 1000 years for the biochar, which meant that 90% of the initial C would remain in the biochar for more than 100 years (BC + 100) subject to the biochar properties and the operating conditions involved in producing it.[50−52] However, the N in the biochar was assumed to convert into heterocyclic compounds, and none of it was assumed to be available for plant uptake and N_2O production.[53,54] The other available nutrients in the biochar contributed to its value as a fertilizer in addition to its other considered benefits such as a 7.2% improvement in fertilizer use efficiency and 50% reduction in N_2O emissions from the fertilizer itself.[55−57] Increased crop yield may be expected on many soils (on average, 15% worldwide and 25% for tropical soils),[58] but it was not within the scope of our analysis as we did not include the selling of crops in our calculations.[51,59] In contrast, since the hydrochar and digestate mineralize rapidly, they were assumed to have a poor performance in carbon sequestration but would provide more nutrients to the soil on the short term, sometimes exceeding the requirement.[60−63] This evaluation of the hydrochar is likely to change with further investigation given the limited number of studies that have been conducted to determine its nutrient release dynamics and availability.

Other Processes. Additional processes involved in our analysis included heat and power co-generation, anaerobic digestion, hydrotreating, biocrude upgrading, transportation, and construction. Several of these are relatively well known, and there was abundant data available to calculate associated parameters, as well as their costs. However, the operating data for some of the processes were sparse, and their parameters were based on several assumptions. All of the processes are described in the Supporting Information with details and assumptions used.

Life Cycle Assessment (LCA). The environmental assessment presented in this study was performed using a cradle-to-grave LCA approach (determined by the system boundaries depicted in Figure 1). LCA is defined as a framework used to analyze the environmental impacts of a product, process, or system throughout its life.[64] It generally involves four main steps, which include goal and scope definition, inventory compilation, impact assessment, and interpretation. Each of these is employed in this study and have been further explained in the following sections. An avoided burden approach was used in which displaced products provided a corresponding environmental benefit and allocation methods were avoided. The various steps involved in conducting an LCA are defined below with additional information.

Defining Functional Unit and System Boundaries. The system included every process in the life cycle starting from poultry litter collection to end-product utilization, as well as all of the associated energy exchange and emissions (Figure 1). The functional unit for this study was defined as the management of 1000 kg of fresh or wet poultry litter with a 25% w/w moisture content (proportion given on a wet basis). The functional unit was based on mass flow, as it was easier to perform calculations and interpret the results derived from the mass distribution of the products.

Life Cycle Inventory (LCI). The LCI plays a key role in an LCA as the basis for the subsequent LCIA, sensitivity analysis, and economic analysis. LCIs associated with the treatment of 1000 kg of wet poultry litter for the six technologies, as well as for the reference case, were compiled (Table S3 in the Supporting Information). It is important to note that we did not perform any process modeling to obtain the necessary initial data for the analysis. The values in the life cycle inventory table were derived from a combination of the mass and energy balances, from the literature and experimental results cited in the Supporting Information, as well as from the Ecoinvent Database.[65]

For the carbon dioxide (CO_2) emission data, we did not include the emissions that were linked with the feed production (Figure 1). This is because we restricted our system boundary to include poultry litter collection only and not the preceding activities involved in rearing the poultry such as photosynthesis, crop planting and growth, feeding of poultry, and production of the litter itself. If we were to expand our system boundaries to include these activities, the CO_2

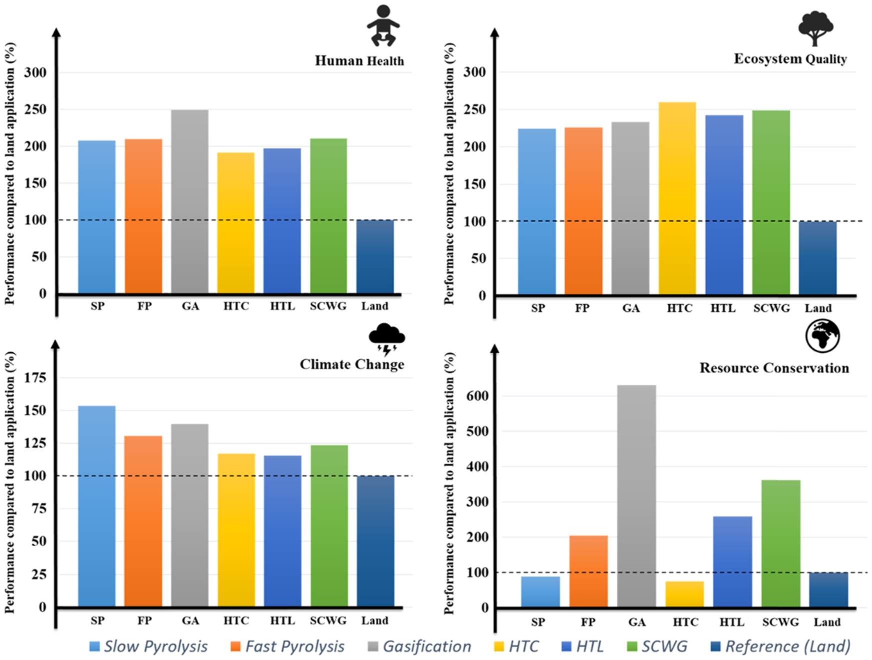


Figure 2. Difference of the net environmental performance (end points) of the six thermochemical technologies (slow pyrolysis (SP), fast pyrolysis (FP), gasification (GA), hydrothermal carbonization (HTC), hydrothermal liquefaction (HTL), and supercritical water gasification (SCWG)) compared to that of the reference case (land) of land application (fixed at 100% for all categories). This diagram has been specifically designed so that a higher proportion corresponds to better environmental performance (and correspondingly lower LCA points). "Resource conservation" in the figure represents the "resource depletion" category and has been provided with an alternative name to avoid misinterpretation.

uptake combined with carbon sequestration in the biochar and other emission reductions could offset all or a part of the CO_2 emissions produced throughout the processes. Therefore, the full life cycle impact of the different pathways on carbon sequestration or greenhouse gas emissions cannot be ascertained from this analysis. However, the comparison between the pathways and the direct application of poultry litter was not affected as the feedstock for all of the cases is considered to be the same.

Life Cycle Impact Assessment. The LCIA was carried out using the IMPACT 2002+ method, which involved 15 midpoint impact categories followed by four end-point (damage-oriented) categories.[66,67] The categories, structure, and weighting factors used were default values associated with the method and are provided in the Supporting Information (Tables S6 and S7). The results were presented at three different levels: midpoints, damage categories (end points), and normalized points. This is because though there is increasing uncertainty as we move to a higher level, the calculation of a single value for each technology made it easier to compare their performance.[67] A single point represents the average impact in a specific damage category caused by a person during 1 year in Europe.[66] These are default values, and though there are studies that provide conversion factors based on different geographic locations, they were missing the factors for some of the categories and hence were not incorporated here.[68]

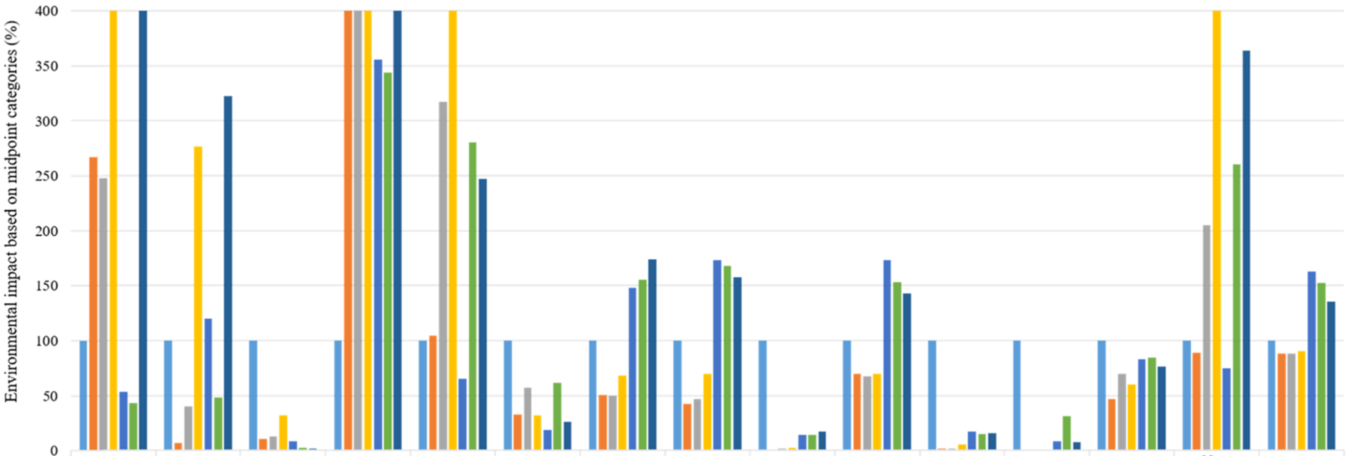
Technoeconomic Analysis (TEA). The main economic components considered in this study were the capital costs and operation and maintenance (O&M) costs of the thermochemical technologies, and the secondary treatment costs (AD, CHP, and refinery plants) and transportation costs of both raw materials and products. A discount rate of 10% was selected for the annual capital cost calculation, and plants were assumed to have a 30 000 kg daily capacity, calculated by equally allocating the estimated poultry waste in New York State to 10 plants.[26,69] The profits from salable products and carbon credits (base scenario of \$20/1000 kg CO_2 equiv) were considered as the sources of revenue.[70] Biofuels, electricity, and heat were all assumed to be sold at the market price of substituted products. The prices of soil products were calculated by using the avoided fertilizers' values, but the non-nutrient values were not considered (which may amount to over 80% of the value of the solid product).[71,72] Details about equations and factors, as well as assumptions made, are provided in the Supporting Information.

Coexistence of LCA and TEA. It is important to note that the system boundaries for both the LCA and TEA were different (with the one for the LCA being broader to consider the environmental impacts of products such as biochar) and they were each meant to evaluate different aspects of the considered systems (environmental impact through the LCA and economic performance through the TEA). There is considerable uncertainty regarding the potential economic benefits of products such as biochar, and hence the TEA was confined to the selling of the products. This approach involving the coexistence of the two methodologies has been employed in other similar studies too.[73,74] One step further would be to integrate both the analysis within a multicriteria objective requiring optimization or to incorporate the economic evaluation of ecosystem services, but that was beyond the scope of this study.[75]

Sensitivity Analysis. The results obtained from the LCA and TEA performed vary considerably based on variations in any of the considered input parameters. The scarce availability of practical plant data, the scaling effects, the learning effects, unfair assumptions, and variation in local conditions could all have impacts on the results. To address these challenges, we estimated the range of important parameter values and conducted a sensitivity analysis. For each technology, the factors with the highest probability of influencing the results were selected, and the updated LCA and TEA results were recorded with the changes in input parameters. Some of these ranges and values were selected based on the mass balance results and the assumed distribution of products (Table S2), while others were derived from the literature cited in Section S3 of the Supporting Information (summarized in Table S5).

■ RESULTS

Comparative LCA Results. For each technology, significant environmental impact was avoided compared to the direct land application case in most environmental categories at each of the three levels: midpoint, end point (damage



	Carcinogens	Non-carcinogens	Respiratory inorganics	Ionizing radiation	Ozone layer depletion	Respiratory organics	Aquatic ecotoxicity	Terrestrial ecotoxicity	Terrestrial acidification	Land occupation	Aquatic acidification	Aquatic eutrophication	Global warming	Non-renewable energy	Mineral extraction
■ Land	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
■ SP	266.76	6.57	10.48	535.20	104.57	32.87	50.43	42.50	1.07	69.67	1.68	1.13	46.62	88.61	88.44
■ FP	247.61	39.87	12.58	501.21	317.27	57.19	49.76	47.11	1.44	67.48	1.97	1.10	69.36	205.05	88.30
■ GA	1521.90	276.80	32.15	2043.56	620.91	31.74	68.34	69.52	2.31	69.60	5.20	1.19	60.45	635.39	90.29
■ HTC	53.73	120.22	8.22	355.66	65.29	18.71	148.08	173.52	14.00	173.27	17.23	8.08	83.00	74.63	162.49
■ HTL	42.95	48.41	2.31	344.25	280.26	61.89	155.32	167.72	14.59	153.11	14.92	30.91	84.52	260.45	152.26
■ SCWG	739.74	322.12	1.47	970.88	246.95	26.19	174.21	157.85	17.23	143.16	15.46	7.32	76.50	363.78	135.48

Figure 3. Calculated LCIA midpoints for the different technologies. Lower values correspond to better environmental performance as we are comparing the proportion of the obtained points for each category.

Table 1. LCIA End Points and Normalized Total Points among Technologies[a]

	human health (pts/kg)	ecosystem quality (pts/kg)	climate change (pts/kg)	resource depletion (pts/kg)	total points (pts/kg)	total points (pts/ton)
reference case	1.24×10^{-4}	1.49×10^{-5}	1.42×10^{-4}	-1.09×10^{-5}	2.71×10^{-4}	0.271
slow pyrolysis	-1.01×10^{-5}	-3.61×10^{-6}	6.64×10^{-5}	-9.63×10^{-6}	4.31×10^{-5}	0.043
fast pyrolysis	-1.26×10^{-5}	-3.82×10^{-6}	9.88×10^{-5}	-2.22×10^{-5}	6.02×10^{-5}	0.060
gasification	-6.10×10^{-5}	-5.02×10^{-6}	8.61×10^{-5}	-6.85×10^{-5}	-4.85×10^{-5}	−0.048
HTC	1.01×10^{-5}	-8.95×10^{-6}	1.18×10^{-4}	-8.21×10^{-6}	1.11×10^{-4}	0.111
HTL	3.10×10^{-6}	-6.33×10^{-6}	1.20×10^{-4}	-2.82×10^{-5}	8.89×10^{-5}	0.089
SCWG	-1.34×10^{-5}	-7.30×10^{-6}	1.09×10^{-4}	-3.93×10^{-5}	4.90×10^{-5}	0.049

[a] Negative values correspond to better environmental performance.

categories), and normalized points (Figures 2, 3, and S4). The reference case total LCIA normalized score was calculated to be 0.271 points (pts)/1000 kg wet poultry litter, while the scores for other technologies were in the range of −0.048–0.111 pts/1000 kg wet poultry litter (Table 1). Among the investigated technologies, gasification performed the best with −0.048 pts/1000 kg wet poultry litter. However, the performance of the technologies in the individual damage-oriented impact categories varied (Table S8 and Figure 4b) and had lesser uncertainty associated with it. Hence, the results were presented at each of the three levels to avoid confusion, and it is essential that the readers understand the differences between them. It is important to note that Figure 2 is designed to portray better environmental performance through higher bars, whereas in Figure 3 and Table 1, we directly display the LCA points where a lower value corresponds to improved environmental performance. Figure 4a provides a contribution analysis in the midpoint performance of slow pyrolysis (similar for all of the other scenarios), whereas Figure 4b goes one step further to include the magnitude of the avoided burdens for representative technologies. The contribution analysis for all of the other technologies is provided in Figure S3 of the Supporting Information.

Human Health Impact Category. Human health was one of the impact categories in which all of the technologies presented a clear and appreciable improvement over the reference case (92–149% better performance) (Table 1 and Figure 2) due to their elimination of the adverse effects of direct land application of poultry litter. The ionizing radiation-midpoint category for slow pyrolysis, linked to the direct and indirect electricity consumption, was the most prominent contributor (18%) in this category (Figure 4). NH_3 emissions from HTL soil products (primarily digestate), when applied to land, appeared to be a critical component (25%) for this impact category based on the breakdown analysis (Figure 4b) and contributed prominently to the respiratory inorganics-midpoint category. However, the value was still lower than that of the reference case, which showed high values of respiratory organics and respiratory inorganics-midpoint categories due to the associated emissions and thus had the worst overall performance (0.124 pts/1000 kg wet poultry litter) (Table 1). Gasification was found to have the best performance (−0.061 pts/1000 kg wet poultry litter), with electricity and heat production avoidance playing a big role (60% contribution) (Figure 4b). However, we must point out that one cannot accurately quantify and identify the exact values of the characteristic factors for human health yet, and there is considerable debate regarding their representation. This should be considered while interpreting these results.

Figure 4. (a) Contribution of factors in the midpoint environmental categories for slow pyrolysis (diagrams for other technologies in SI); (b) breakdown analysis for the absolute life cycle impact on human health, ecosystem, climate change, and natural resource categories for slow pyrolysis (SP), hydrothermal liquefaction (HTL), and gasification (GA) in comparison to the reference case of direct land application (land). Any labels below the *x*-axis refer to emissions or consumption. Labels in brackets above the *x*-axis refer to avoided products/resources. The points on the *y*-axis refer to the negative of the normalized values of the four end-point categories so that we can present positive environmental impacts above the *x*-axis and negative impacts below the *x*-axis to make it easier to interpret the results. "Resource conservation" in the figure represents the "resource depletion" category and has been provided with an alternative name to avoid misinterpretation.

Ecosystem Quality Impact Category. All of the technologies outperformed the reference case in terms of impacts on ecosystem quality (improvement of 0.0185 pts or more), but their results compared to each other were very similar and within a narrow range of 35% (Figure 2). The hydrothermal technologies performed better than gasification, which in turn was better than the pyrolysis technologies (Table 1). Nutrient flow seemed to be the most important contributor, with avoided fertilizers providing high benefits but nutrient leaching and associated pollution having the maximum negative impact. As an example, the N and P runoff for the reference case had a combined effect of 85% on the final score of 0.015 pts/1000 kg wet poultry litter (corresponding to 205 PDF m^2 year) (Table 1 and Figure 4b). Surprisingly, this category only contributed in the range of 4−10% to the overall point calculations for all of the technologies, and this could be attributed to both its normalization factor (Table S7) and the characterization factors for its midpoint categories such as aquatic and terrestrial ecotoxicity, land occupation, and aquatic eutrophication provided by the LCIA methodology (Table S6).

Climate Change Impact Category. This category had the highest influence on the overall points' calculation for each technology (39−79% contribution). An improvement of 15−53% was provided by the technologies in this category compared to that of the base case (0.142 pts/1000 kg wet poultry litter), although their individual performances varied, with slow pyrolysis (0.0638 pts/1000 kg), fast pyrolysis (0.09877 pts/1000 kg), and gasification (0.08608 pts/1000 kg) outperforming the hydrothermal technologies (0.109−0.120 pts/1000 kg) (Table 1 and Figure 2). The life cycle net GHG emissions per functional unit for slow pyrolysis, gasification, and HTL based on the points were calculated to be 657 kg, 852 kg, and 1191 kg CO_2 equiv/1000 kg wet poultry litter, respectively, compared to the 1410 kg CO_2 equiv emissions from the reference case (Table S8). From the breakdown analysis, we noted that CO_2 emissions contributed the most to the climate change impacts for the three representative technologies (65−71% of total points for the category) (Figure 3). Significant GHG emission abatement from slow pyrolysis in comparison to the reference case was observed in the amount of −753 kg CO_2 equiv/1000 kg wet poultry litter, highlighting the benefits of the carbon storage function in the biochar (−728 kg CO_2 equiv/1000 kg wet poultry litter). Though not as good as slow pyrolysis, gasification had better environmental performance than HTL owing to the avoidance of external electricity production and the HTL's hydrochar lacking any carbon storage capability. The gasification system also outperformed fast pyrolysis in terms of climate change impacts (Figure 2). The heat and electricity produced from the syngas can replace a considerable amount of fossil-based heat and electricity that contributes to its better overall environmental performance. In comparison, the bio-oils produced from fast pyrolysis also help avoid fossil-based emissions, but the upgrading of the oil leads to additional environmental impacts.

Resource Depletion Impact Category. Gasification (−0.0685 pts/1000 kg wet poultry litter) performed favorably in the resource depletion impact category by a large margin (530% improvement) compared to the reference case (−0.0109 pts/1000 kg wet poultry litter), as well as the other technologies (Table 1 and Figure 2). For slow pyrolysis, HTL, and gasification, the net primary energy saved for 1000 kg of wet poultry litter was calculated to be 1464, 4286, and 10 412 MJ, respectively, in comparison to that of the 1652 MJ avoided through the reference case (Table S8 in the Supporting Information). A significant amount of heat and electricity was generated from gasification, which was more than the energy consumed throughout the life cycle (83% contribution to results) (Figure 4b). As opposed to gasification, the resources saved from slow pyrolysis and HTC were lower than that of the reference case. Even though the biochar improved soil fertility, none of the N in the biochar made by slow pyrolysis, fast pyrolysis, and gasification was considered available for plants to utilize as in contrast to the hydrochar, digestate, and poultry litter itself, which were assumed to avoid more fertilizer production owing to the rapid release of nutrients.

TEA Results. The gasification process had the highest revenue per 1000 kg wet poultry litter among any of the conversion technologies, owing to electricity ($120) and heat ($114) production (represented by "CHP revenue" in Figure 5) but the costs for plant operation ($155/1000 kg wet poultry litter), biogas transportation, and CHP ($68/1000 kg wet poultry litter, including gas clean-up), were found to offset this benefit. However, the cost for the pyrolysis plants ($45 and $76/1000 kg wet poultry litters for slow and fast pyrolysis, respectively) was only about half the revenue from the biochar ($100/1000 kg wet poultry litter for slow pyrolysis) and 85% of the revenue from gasoline and diesel produced through biocrude ($89/1000 kg wet poultry litter for fast pyrolysis). The wide range for pyrolysis stemmed primarily from the variable biochar value, where a mature market has not been formed yet. HTL had the best economic performance (−$46.3/1000 kg wet poultry litter) because of its low O&M costs ($52/1000 kg wet poultry litter) and the utilization of existing ADs, even though its products' total revenue ($143/1000 kg wet poultry litter) was lower than that of slow pyrolysis ($168/1000 kg) and gasification ($301/1000 kg). If AD has to be built, the costs increased by 15% (new overall value of −$60/1000 kg wet poultry litter), which was comparable to the economic performance of slow pyrolysis (−$67/1000 kg wet poultry litter).

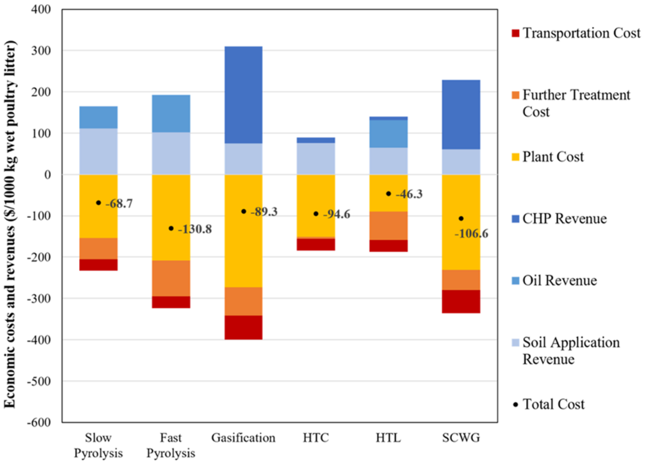


Figure 5. Economic performance of the six technologies. The black points represent the net revenue/1000 kg wet poultry litter in the base-case scenario. Under the base scenario assumptions, none of the six technologies can attain revenues per 1000 kg of wet poultry litter feedstock without sufficiently valuing the environmental performance or the biochar.

The economic performance of the thermochemical technologies was all worse than that of the reference case of direct land application of poultry litter (which would only include the transportation components and some saved fertilizers) based on current assumptions of 50 km transportation and a \$182/1000 kg value of the biochar.

In this study, the breakeven prices calculated based on major revenue sources for slow pyrolysis, fast pyrolysis, and gasification were approximately \$0.31/kg biochar, \$0.055/MJ gasoline/diesel, and \$0.17/kWh electricity produced, respectively. These values were considerably higher than those considered in this study (\$0.182/kg biochar, \$0.024/MJ gasoline/diesel, and \$0.12/kWh electricity) though the value of the biochar varies considerably based on the feedstock. Compared with other biomass-fed technologies, the capital and O&M costs were higher, but the costs associated with the feedstock were much lower (embedded in the transportation cost that varied from \$28 to \$58/1000 kg wet poultry litter). As compared to this, the average cost of land applying poultry litter is in the range of \$12−25/ton.[76] It is important to note that we did not include the cost of raising poultry, as well as the associated resources, CO_2 uptake, and emissions, in our study. While all seven cases can be compared for those steps that they differ in, the comparisons with other studies have to be made with caution since they may or may not include poultry operation itself in the system boundary.

Sensitivity Analysis Results. For slow pyrolysis, LCA results were sensitive to, and thus to some extent driven by, the following inputs: biochar effective year (−40 to +61% change in output LCIA points), electricity consumption (−79 to +60%), as well as the product yields (Figure S5). It appeared that both the biochar and biocrude yields had a greater effect on decreasing the values in the LCA than increasing them owing to the nonlinearity involved in the relationship, but this is highly dependent on the input range selected and the base case used for calculations (Table S5). Transportation distance had negligible effects (−7 to +7%) on the LCA results for slow pyrolysis, while, on the other hand, it was an influential factor (−38 to +41%) in the TEA sensitivity analysis results along with the biochar value (−146 to +95% change). However, the CO_2 price was found to have the largest effect (−19 to +484% change) on the TEA results for slow pyrolysis, with an expected profit of \$264/1000 kg wet poultry litter at a price of \$500/1000 kg CO_2 equiv. The NO_x removal efficiency was also varied from 0.7 to 0.99 and produced a change of +30 and −7%, respectively, in the revenue generation.

For HTL's LCA sensitivity analysis, the land application emissions dominated (−52−13% change in output). However, this is a factor that cannot be easily controlled or clearly investigated and thus always results in high variability within the LCA results. Both the hydrochar yield (−23 to +43% effect) and biocrude yield (−15 to +26% effect) were influential in the sensitivity results for LCA and even more so for the TEA results, with a change of −57−107% and −43 to +126% for the revenues based on variations in the hydrochar and biocrude yields, respectively (Table S5 and Figure S5).

In contrast to the slow pyrolysis and HTL results, where product yields and nutrient flow were prominent factors, the energy flow played the most critical role in both the LCA and TEA sensitivity analysis results of the gasification system. A change of −19 to +102% in LCA points and −79 to +13% in revenue generation was observed for a change in the CHP efficiency from 0.6 to 0.95.

A common trend observed with all of the three systems (slow pyrolysis, HTL, and gasification) was that the LCA results were insensitive to the transportation distance (0.093, 0.063, and 0.028% changes in LCIA points per change in the distance for a fixed mass of the biochar transported, respectively), whereas the TEA results were considerably affected (change of 0.52% in revenue per change in the distance for a fixed mass of the biochar transported for both slow pyrolysis and HTL) (Figure S5).

DISCUSSION

Environmental Impact of Thermochemical Technologies. While the environmental performance of all six thermochemical technologies was superior to that of the reference case of direct land application of poultry litter by a large margin, the recovered resources could not outweigh the upstream energy/resource consumption or the emissions induced for most of the technologies in our base-case scenario. Gasification technology's favorable performance in the LCA for our base-case scenario could be attributed to its high energy recovery.[77] The biochar from the pyrolysis technologies greatly aided its performance due to its multiple benefits, even though the energy embedded within it was not used.[51] On further analysis, we found that if the entire biochar produced was to be used for energy production, the environmental performance of that system (0.124 pts/1000 kg wet poultry litter) would be three times worse than the system involving soil application of the biochar as we considered in our base-case scenario (0.043 pts/1000 kg wet poultry litter). This was the case without consideration of additional benefits of soil application of the biochar such as crop yield increase, increased soil organic carbon, and water retention capacity, which were not within the scope of our analysis and would further improve the environmental performance of the system.[51,52] For the hydrothermal technologies, the results were highly dependent on the pathways chosen for their multiple product fractions, and the hydrochar and the digestate, with their high-nutrient contents, were prime examples of this. Therefore, nutrient recycling was a major environmental benefit, and its contribution was either comparable to or greater than that of the greenhouse gas emission reductions and energy generation in almost every impact category.

Economic Feasibility of Thermochemical Technologies for Poultry Litter Conversion. Based on the TEA calculations, \$68.7, \$130.8, \$89.3, \$94.6, \$46.3, and \$106.6 per 1000 kg of wet poultry litter for slow pyrolysis, fast pyrolysis, gasification, HTC, HTL, and SCWG, respectively, were the minimum amount of additional revenue required to attain a net positive value at the base scenario. However, this would only hold true if we were to assume that all of the other costs and prices remained constant, and that is highly unlikely. The sensitivity analysis helped identify the important parameters for both the analyses, but it is important to note that there would be additional variabilities introduced in the systems based on local conditions such as distances, prices, effects of soil products, and so on. As an example, the performance of the biochar in terms of fertility and crop yield improvement could be very different in places with weathered soil as compared to that of the productive soil found in the U.S. corn belt.[51,56] Furthermore, a crop yield improvement of 28% through poultry litter biochar[56] could have a major impact on the economic performance of the pyrolysis technologies and results in a positive NPV of +\$790/1000 kg wet poultry manure for our considered base scenario of the slow pyrolysis system using a value of \$1741/1000 kg dry dairy manure biochar.[72] This should be further analyzed and verified in future studies.

Policy Implications and Future Research Directions. Even though the thermochemical technologies proved to be environmentally attractive, the high capital and O&M costs involved made it difficult to achieve profitability without an additional income from sales of biochar, crop yield increase, and carbon trading, and suggested the need for biochar market development as well as stronger government regulation through subsidies and internalizing cost for environmental damages. Financial incentives for valuable "green" products such as biochar, biocrude, and the generation of cleaner electricity and heat need to be provided to encourage the introduction of these technologies at large scales. Additionally, with increasing public attention on waste recycling technologies, more financial benefit mechanisms should be established for nutrients and waste recovery products. This would provide a considerable improvement in the economic performance of the thermochemical technologies. Other factors aiding their economic performance in the future could include (1) lower capital costs as the industries producing the equipment become more commercially mature; (2) an evolved system, allowing centralized production, and large-sized plants, which would reduce unit operation costs; and (3) carbon trading opportunities becoming more supportive in the future, with an expected range of at least \$40−80/1000 kg CO_2 equiv to stay consistent with the Paris Agreement temperature goal, and some studies estimating prices of \$100−1000/1000 kg CO_2 equiv based on different scenarios and timelines.[70,78,79] These potential improvements were not included in the current assessment. As an example, an increase in revenue by \$58.21/1000 kg wet poultry litter for slow pyrolysis and \$27.72/1000 kg wet poultry litter for fast pyrolysis could be achieved at \$100/1000 kg CO_2 equiv in our current analysis. A carbon price of \$127/1000 kg CO_2 equiv was found to be the breakeven value for slow pyrolysis. Implementation of such a technology would then also more than double the human health benefits as well as ecosystem quality beyond climate change (such as water quality), as shown in this analysis.

Under the current policy in the United States, where only very low environmental credits are granted that are typically limited to greenhouse gas emission reductions, a financially feasible solution will be highly dependent on the trade-off between the scale of plants and transportation distances. Plant scale-up would considerably lower unit plant costs and help increase yields and efficiency. However, both the supplier and the various markets involved would then be further away from the plant, increasing transportation distances and their associated costs and required infrastructure networks. In addition, any long-distance transport imposed by possible regulations that discourage nearby disposal of poultry litter or by the need to close nutrient loops as part of a circular economy may reduce transportation costs of wet poultry litter by 56% from \$363 to \$160 per 100 km for our considered plant capacity (30 000 kg wet poultry litter per day). One way to overcome these challenges could be the utilization of supply-chain optimization along with the spatial analysis to design integrated centers for processing different waste feedstocks from distributed farms, thus allowing for larger capacities and aggregated management. Furthermore, by adjusting the mix of incoming waste streams, we could ensure that the composition and volume of wastes entering the plant are optimal. Introducing these enhancements in future studies would provide additional insights as to the best choices to make. Our results also suggested that a detailed investigation of further treatment and end use of the primary products should be carried out, as it could lead to additional economic and environmental benefits or burdens not accounted for in the current work. Improvements such as a lower production cost and higher yield ratios for desirable products could be achieved in the near future, and these factors should be assessed in emerging technologies at different scales of implementation.

■ CONCLUSIONS

This study highlighted the potential environmental benefits that thermochemical technologies could provide in comparison to that of the conventional land application of poultry litter through the LCA. A framework to analyze both the economic and environmental performance of these technologies was also developed so that they could be compared with the existing alternatives such as anaerobic digestion and incineration that are more mature and have already been researched in detail. While the current technoeconomic analysis was based on fixed base-case values, the sensitivity analysis served as an important reminder that there is a lot of uncertainty and variability associated with the economic parameters for these novel technologies and that certain policy decisions such as carbon credits could have a huge impact on the results. Our analyses allow prioritizing future empirical studies to reduce uncertainties in information on energy generation, the length of biochar soil effects, and the product distribution from the various technologies considered.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.0c02860.

Description of thermochemical technologies, LCI data, and assumptions; LCIA methodology, categories, and factors; technoeconomic analysis parameters, sensitivity analysis parameters, and results; and additional LCA results (PDF)

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Notes

The authors declare no competing financial interest.

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