



Reducing the environmental impacts of plastics while increasing strength: Biochar fillers in biodegradable, recycled, and fossil-fuel derived plastics

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ABSTRACT

In this study, material characterization and a cradle-to-gate life cycle assessment methodology are applied to examine how the addition of biochar as a filler in recycled plastics (rHDPE) influences material properties and environmental burdens. Environmental impacts for these composites are then compared to biochar fillers in virgin HDPE and bio-based polylactic acid (PLA) and polyhydroxybutyrate (PHB). At 40% biochar addition to rHDPE, biochar increased the tensile strength by 45%, stiffness by 126% and flexural storage modulus by 79% but resulted in a more brittle material. Biochar addition detrimentally affects the thermal degradation of both PLA and PHB, resulting in reduced mechanical properties. Increased biochar loading linearly reduced the global warming potential of all plastics relative to neat plastic by up to 3.3 kg CO₂ eq. per kg of composite, with a dual benefit of reducing the amount of plastic used and creating a net-uptake of carbon in the biochar. Similarly, biochar decreased the fossil fuel depletion of PLA, PHB, and HDPE, but additional transportation-related emissions and the low fossil fuel depletion for neat rHDPE caused biochar addition to increase fossil fuel depletion for rHDPE. While changes in material application and end-of-life were outside the scope of this study, biochar is expected to provide further end-of-life benefits to the biodegradable PHB or PLA. Importantly, this study demonstrates that biochar can be applied to produce carbon-neutral composite materials when added to a wide variety of plastics and demonstrates the potential of biochar to reduce the environmental impacts of plastic materials.

1. Introduction

Plastics are one of the most ubiquitous materials in modern society, but their production is highly dependent on petroleum resources, and they are currently responsible for notable environmental impacts [1–3]. As the use of plastic products continues to grow, forecasts indicate that plastic production will be responsible for more greenhouse gas emissions than coal by 2030 [4] and the majority of the growth in global fossil fuel use by 2050 [1]. To address these challenges, new approaches are needed to mitigate these impacts and reduce reliance on fossil resources during the manufacturing of plastic products.

One strategy proposed to reduce the negative environmental impacts of plastics is the addition of biochar as a filler material in the plastic. Biochar is a high carbon-content material produced by pyrolysis of biomass and has been extensively studied as a method to sequester carbon [5–7]. The material properties of biochar vary based on its feedstock and processing; with optimal processing, biochar can be produced with comparable properties to standard fossil fuel-derived carbon

filler materials (e.g., carbon black) [8–10]. Because of these beneficial properties, biochar has been widely studied as a filler material in plastics. Typically, the addition of biochar improves the strength and stiffness of fossil-fuel-derived plastics across a wide variety of plastic types and biochar feedstocks [11–20]. For example, a recent study found that addition of 50 wt.% rice husk-derived biochar to HDPE results in a 146% increase in flexural strength, a 200% increase in tensile modulus, an 11% increase in tensile strength, and a 40% increase in tensile modulus [11]. In contrast, the addition of biochar to biodegradable plastics (e.g., PLA or PHB) reduces the thermal stability of the material, resulting in decreased mechanical properties of the biodegradable plastics [21–28].

Numerous life cycle assessments have examined the environmental impacts of wood-plastic composites (WPCs) and shown the relative GHG reductions compared to neat petroleum-based plastic polymers [29–32]. Despite the environmental benefits of WPCs, various limitations include inferior mechanical properties and dimensional instability, which could reduce the service life of these materials, leading to potentially higher environmental impacts due to product replacement [33]. Biochar offers

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numerous mechanical and environmental benefits over wood, including increased flexural and tensile strength [15,33,34], utilization of a wider variety of waste materials, and the propensity to sequester carbon for a longer period of time due to biochar's high chemical stability [35]. Past life cycle assessment studies have found that biochar can reduce certain environmental impacts of plastic products relative to petroleum-derived filler materials [36–38]. However, these studies have been limited in scope to automotive products and have not examined waste-derived biochar (such as biochars from agricultural or forestry waste that have been commercialized for soil amendment and carbon sequestration). Further, these studies have only examined biochar addition to polypropylene. The environmental impact of biochar addition to other petroleum derived plastics and recycled or biobased plastics remains a crucial gap to understanding how biochar can be utilized to drive reductions in the environmental impacts of plastics and their products.

In this study, we conduct both mechanical property and environmental impact comparisons between recycled high-density polyethylene (rHDPE) with and without biochar filler. Specifically, tensile and flexural properties are assessed with varying filler levels, and we apply a cradle-to-gate environmental impact assessment methodology to examine the environmental impacts of biochar as a filler material. To draw comparisons and elucidate additional environmental benefits that could be achieved with biochar as a filler, its application in two biodegradable plastics (polylactic acid (PLA) and polyhydroxybutyrate (PHB)), and a fossil-fuel derived plastic (HDPE) are also examined through environmental impact assessments. However, experimental analysis of the material properties of these additional composites is outside the scope of this work. We hypothesize that the addition of biochar would decrease the global warming potential, as well as other environmental impacts, for all non-recycled plastic types by replacing virgin material with a waste-derived material. Given the recent, rapid growth in interest in applying biochar as a filler material in plastic, this study builds a vital platform to begin to understand how biochar can be applied to reduce the environmental impacts of a diverse set of plastic products.

2. Methods

2.1. Experimental methods

2.1.1. Composite production

Composites of biochar and rHDPE were produced to examine the impact of biochar addition on plastic mechanical behavior. Pacific Biochar (Santa Rosa, CA, USA [39]) supplied wood-derived biochar (85% carbon with an O/C molar ratio of 0.21) produced from forestry residues. Prior to extrusion, this biochar was ball milled for 2.5 min at 30 Hz (1800 rpm) in a Retsch Mixer Mill 400 to reduce particle size, a ball milling procedure which has been previously shown to reduce biochar particle sizes to $<10\ \mu\text{m}$ [8] and dried at $105\ ^\circ\text{C}$ to reduce moisture content. Recycled high-density polyethylene (rHDPE) was provided by Northwest Polymers (Moalla, OR, USA). To demonstrate the impact of Pacific Biochar on the thermal degradation of biobased plastics, PLA was supplied by NatureWorks (2003D, Minnetonka, MN, USA), and PHB was supplied by Tianan (ENMAT Y3000P, Ningbo City, China).

Composite materials of rHDPE with 0%, 20%, and 40% biochar addition were prepared to represent a range of biochar addition (no, medium, and high amounts). Composites were compounded in a Thermo Fisher Scientific HAAKE Minilab II dual screw extruder for 5 min, at a speed of 100 rpm and a temperature of $190\ ^\circ\text{C}$. Rheology data were collected during extrusion to verify sample mixing and consistent biochar addition between batches (Supplemental Figure S1). Samples were extruded into a Thermo Fisher Scientific Minijet Pro-injection molder and injected into Thermo Fisher Scientific DMA three-point bending sample mold (Part #557–2295, $10\ \text{mm} \times 60\ \text{mm} \times 1\ \text{mm}$) or Thermo Fisher Scientific ASTM D638 Type V tensile test mold (Part #557–2299). Samples were injection molded with a $190\ ^\circ\text{C}$ barrel

temperature and a $60\ ^\circ\text{C}$ mold temperature. An injection pressure of 750 bar was applied for 10 s, followed by a post pressure of 450 bar for 60 s. Samples were produced in triplicate for both bending and tensile tests.

2.1.2. Composite testing

The impacts of biochar on the thermal degradation temperature of rHDPE, PLA, and PHB with 0%, 20%, and for rHDPE, 40% biochar addition were measured with thermogravimetric analysis (TGA) on a TA Instruments Q5000 IR, with 20 mg samples. Samples were heated from $30\ ^\circ\text{C}$ to $900\ ^\circ\text{C}$ at a rate of $10\ ^\circ\text{C}/\text{min}$. Differential scanning calorimetry (DSC) was used to measure shifts in the melt peak and melt enthalpy on injection molded rHDPE samples with 0%, 20%, and 40% biochar addition on a TA instruments Discovery DSC 2500. Samples were heated from $-80\ ^\circ\text{C}$ to $250\ ^\circ\text{C}$, then cooled to $-20\ ^\circ\text{C}$ at $10\ ^\circ\text{C}/\text{min}$. Sample crystallinity was determined based on the ratio of sample melt enthalpy to 100% crystalline melt enthalpy ($293\ \text{J/g}$ for HDPE) [40].

The tensile and flexural strength and modulus of the biochar-filled plastics were examined for changes to strength as a function of filler content. A three-point bending fixture measured the flexural dynamic modulus with a TA Instruments Q800 dynamic mechanical analyzer (DMA). Samples of HDPE and 0%, 20%, and 40% biochar were heated from $30\ ^\circ\text{C}$ to $100\ ^\circ\text{C}$, with a frequency of 1 Hz, amplitude of $20\ \mu\text{m}$, and a force track of 125%. Tensile strength and modulus were measured on an MTS C43 load frame with a load rate of $5\ \text{mm}/\text{min}$ and a gage length of 35 mm from clamp to clamp. All DMA and tensile tests were measured in triplicate for each sample type.

2.2. Environmental impact assessment methods

Environmental impact assessments were conducted in four stages: goal and scope, life cycle inventory analysis, impact assessment, and interpretation of results. The goal of this assessment was to examine how plastics with different origins are impacted by biochar addition. Four plastics (rHDPE, HDPE, PLA, and PHB) were analyzed with 0, 20, and 40 wt.% biochar-filler on a cradle-to-gate scope, with use phase and end-of-life being excluded. This system boundary is depicted in Fig. 1. The inventory flows for this model included the material flows and energy requirements associated with bio-based and petroleum plastic production, plastic and biochar extrusion and molding, and transportation impacts for a functional unit of 1 kg of material (plastic with or without biochar filler). The PLA production inventory was based on data from NatureWorks, which is the largest producer of PLA in the US [42]. The inventory for PHB was based on values from Miller *et al.* [43] due to the lack of available industry data. The carbon feedstocks for PLA and PHB were assumed to be corn and sugarcane, respectively. The inventories for HDPE and rHDPE were based on data from a US LCI database [44]. The electricity grid for all plastic production inventories was based on the 2015 US Energy Information Association (EIA) electricity grid data [45]. The inventory for biochar was based off of a study by Peters *et al.* [46]. However, changes were made to account for the fact that biochar is a waste residue rather than a by-product of poplar cultivation. In addition, unlike in the inventory from Peters *et al.*, a credit was not applied to the syngas for the displacement of natural gas. Detailed inventories for each of the bioplastics as well as the biochar are provided as Supplemental File S2. The inventory for the injection molding process was based on the NREL US LCI database, but a California electricity grid mixed was used [44]. The inventory for transportation impacts were derived from Kamau-Devers and Miller 2020 [32].

For this work, biochar was modeled as being produced from biomass residues, and therefore no environmental burdens associated with biomass production were allocated to the char. However, it was assumed that the organic carbon present in the biochar is representative of the carbon dioxide (CO_2) taken up during photosynthesis. The amount of CO_2 taken up by the biochar was calculated using Eq. (1) (where C_b is the carbon content in biochar (kg C/kg biochar)).

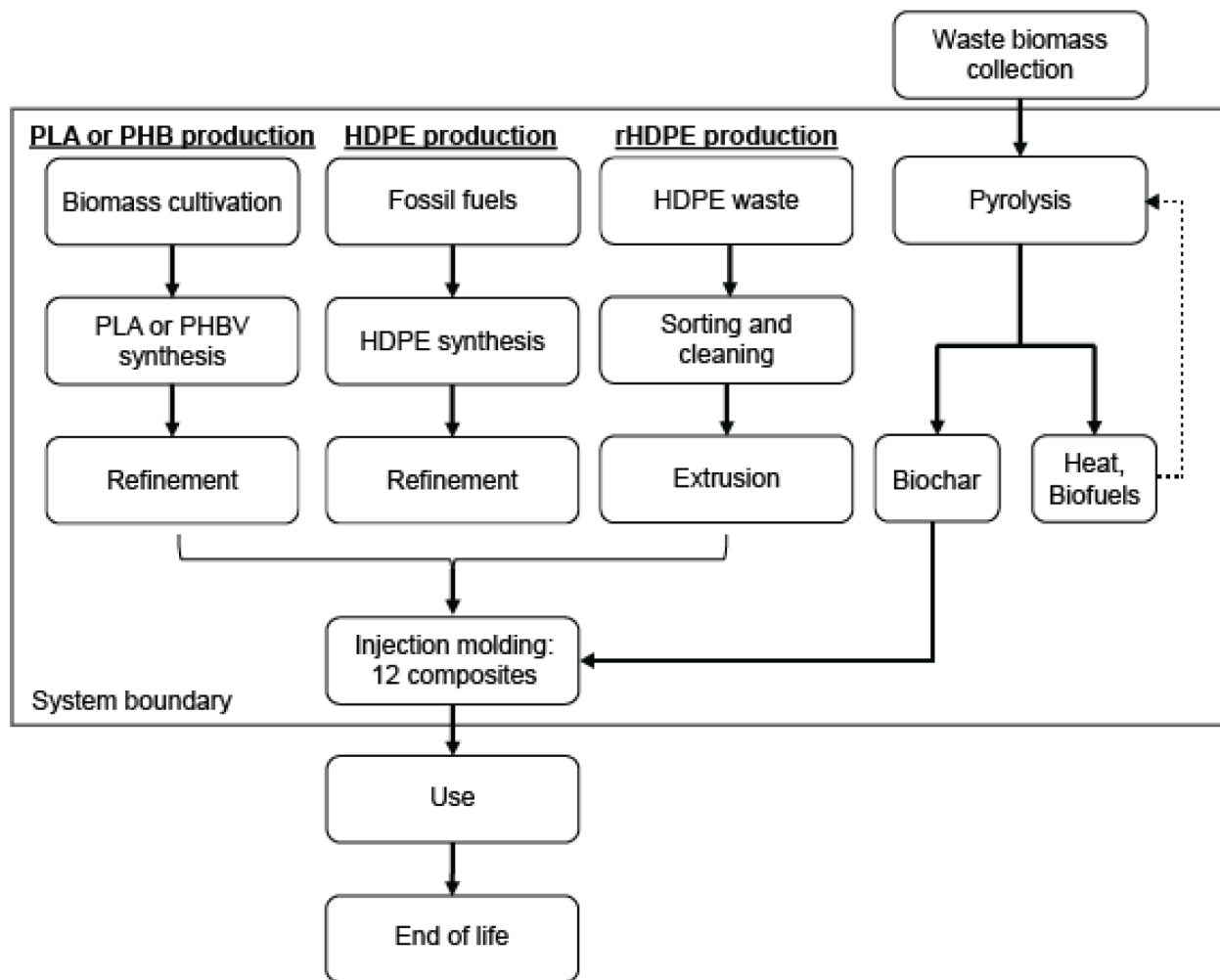


Fig. 1. System boundary of this cradle-to-gate study.

$$CO_2 \text{ stored} = C_b * \frac{1 \text{ mol C}}{12 \text{ kg C}} * \frac{1 \text{ mol CO}_2}{1 \text{ mol C}} * \frac{44 \text{ kg CO}_2}{1 \text{ mol CO}_2} \quad (1)$$

The heating requirements for the pyrolysis process were assumed to be met using the gas and heat resulting from the pyrolysis process itself, therefore negating the necessity for external heat sources such as natural gas [46]. It was assumed that the biochar and plastics were each pelletized before being compounded together. The pelletization of the plastics was part of the inventories selected, and the pelletization of the biochar was modeled based on pelletization following extrusion melt blending with plastic [47]. The plastics and biochar were modeled as being combined via injection molding. If biochar and plastic were prepared at the same location, biochar addition and plastic pelletization could be combined as a single-step process, improving the overall efficiency of the process [44]. The injection molding requirements for bioplastics with and without biochar were assumed to be the same given the negligible difference in heating requirements. Transportation assumptions were derived from Kamau-Devers and Miller 2020 [32]. The plastics and biochar were assumed to travel 200 km and 3340 km respectively to the composite manufacturing site.

The characterization factors from the Tool for the Reduction and Assessment of Chemical and other Environmental Impacts (TRACI) from the US Environmental Protection Agency were used for the environmental impact assessment [41]. This impact assessment method differs from others in that it utilizes input parameters based specifically on US data. This method was chosen due to the abundance of US-based data in the life cycle inventories of the materials analyzed. The interpretation of

results included an uncertainty analysis, which was conducted using input parameters from Kamau-Devers and Miller 2020 [32] who used a quantitative uncertainty method based on pedigree matrices [48,49]. These uncertainties were modeled using distributions for each input, and Monte Carlo simulations were run to determine distributions of anticipated environmental impacts ($n = 10,000$). Median outputs from these simulations are presented in the results.

3. Results

3.1. Experimental results

Addition of biochar to rHDPE had a minimal effect on the thermal degradation temperature of rHDPE (Fig. 2a). Only limited ($<3^\circ\text{C}$) shifts were seen in both onset and peak thermal degradation temperatures for 20% and 40% biochar addition relative to neat rHDPE. Small mass losses ($<2\%$) were observed in both 20% and 40% biochar samples prior to the onset of rHDPE thermal degradation, which are attributed to increased moisture absorption with the addition of biochar, as has been observed in past studies examining biochar addition to plastics [21]. Similarly, addition of biochar has a minimal impact ($<1^\circ\text{C}$) on the melting and crystallization temperatures of rHDPE (Fig. 2b). The glass transition temperature of rHDPE and its composites were not measured due to the typically low glass transition temperature of HDPE ($<-80^\circ\text{C}$). In contrast, biochar addition had a meaningful impact on the crystallinity of rHDPE - as biochar loading increases from 0% to 40%, crystallinity increased from 54% to 81% (Supplemental Table S1). This increase in

crystallinity is consistent with what has previously been observed for addition of biochar to petroleum-derived plastics [15,27]. TGA measurements of PLA and PHB with biochar addition highlight the challenges of applying biochar as a filler material in these plastics. In both cases the addition of 20% biochar reduces the peak temperature of thermal degradation, by 7 °C in PHB and by 23 °C in PLA (Fig. 2c). This decrease has previously been widely observed in studies examining biochar and PLA or PHB composites and has been attributed to alkali and alkaline earth metals (e.g., Na or Ca) catalyzing the thermal degradation of PLA or PHB, resulting in reduced polymer chain length and poor mechanical properties [22,25,27,50]. Therefore, the mechanical properties of biochar-filled PLA or PHB are not examined in this study.

The tensile behavior of the rHDPE was notably altered by the inclusion of biochar (Fig. 3). The addition of biochar at both 20% and 40% levels increased the tensile strength and stiffness of rHDPE. The tensile strength increased from an average strength of 22.19 ± 0.81 MPa for neat rHDPE to 28.06 ± 0.88 MPa at 20% biochar addition and to 32.29 ± 0.70 MPa at 40% biochar addition, with variations representing one standard deviation between the three measured samples. A similar trend is seen in the tensile modulus, with average Young's modulus values and standard deviations of 606 ± 2.50 MPa, 859 ± 81.7 MPa, and 1371 ± 51.8 MPa at 0%, 20%, and 40% biochar addition, respectively. Along with these increases in strength and stiffness, the addition of biochar modified the failure behavior of rHDPE. For the neat rHDPE, after the peak in tensile stress ($\epsilon = 0.07$ mm/mm), a period of strain softening is observed (from $\epsilon = 0.07$ to 0.4 mm/mm), followed by a period of mild strain hardening until failure, consistent with expected failure behavior for rHDPE [51]. In contrast, samples with 20% biochar addition fail immediately following the strain softening phase, and samples with 40% biochar addition exhibit brittle failure immediately following the peak in stress. Accompanying this greater degree of brittleness with biochar is a decrease in strain to failure, from 2.41 ± 0.30 mm/mm at 0% biochar addition to 0.089 ± 0.015 mm/mm at 40% biochar addition. The improvements in strength and stiffness of rHDPE with the addition of biochar are consistent with past studies examining biochar addition to HDPE and other fossil fuel-derived plastics [11,12].

Like the tensile results, flexural DMA results show that increasing biochar content increases the loss and storage moduli of rHDPE (Fig. 4). At low temperatures (30 °C), the differences between 0%, 20%, and 40% biochar addition are small relative to higher temperatures, with an increase of 43% and 79% of the storage modulus of neat rHDPE for 20% and 40% biochar addition, respectively. Similarly, relatively minor differences are seen in the loss modulus at lower temperatures. As temperature increases, differences between storage modulus at 0%,

20%, and 40% biochar content also increase, indicating that the addition of biochar may also improve the heat deflection temperature of rHDPE. The tangent of delta decreases with increasing biochar content, indicating a more elastic response, a finding consistent with the increase in elastic modulus observed via tensile testing.

3.2. Life cycle assessment result

For all plastic types, the addition of biochar decreases the global warming potential of the material, with a linear relationship between the amount of biochar added and the reduction in global warming potential (Fig. 5a). The addition of biochar to plastic materials provides a dual benefit of reducing the global warming potential associated with plastic production and creating a net uptake of CO₂ in the biochar. This dual benefit results in approximately twice the percent reduction in GWP as the percent addition of biochar (e.g., a 20% biochar addition results in an approximately 40% reduction in global warming potential). The highest percent change is noted for rHDPE, which has limited GWP associated with plastic production, and for which the utilization of a biochar filler at 40% suggested the potential for net-uptake composites. In contrast, PHB had the lowest percentage change as it has the highest GWP associated with plastic production and, as a result, maintained relatively high global warming potential even with biochar filler. For most plastic types, a similar trend to GWP is seen for fossil fuel depletion (Fig. 5b). However, rHDPE shows a negligible increase in fossil fuel depletion with biochar addition; this result is a function of the production of the material, which requires little fossil fuels relative to the additional transportation step. The addition of biochar to PLA, rHDPE, and HDPE contributes to limited change in acidification and respiratory effects (Figs. 5c&d). However, in PHB, the addition of biochar results in decreases in these impact categories. This decrease is due to the relatively large acidification and respiratory effects associated with PHB production relative to PLA or petroleum-derived plastics [43]. Other environmental impact indicators (ozone depletion, smog formation, eutrophication, human health cancer, human health noncancer, and ecotoxicity) were examined (Supplemental Figure S3) and showed similar trends to those of acidification and respiratory effects.

For all plastics except rHDPE, the magnitude of the impact is dominated by the production of the plastic (Fig. 6). This constituent production makes up over 70% of the global warming potential and 60% of the fossil fuel depletion for all percent biochar additions, with the percentage increasing with increasing biochar content. Similar trends are seen for acidification, respiratory effects, and other impact indicators (Fig. 6 and Supplemental Figure S4). Conversely, in rHDPE, this relationship is not present as the sorting and recycling process requires less

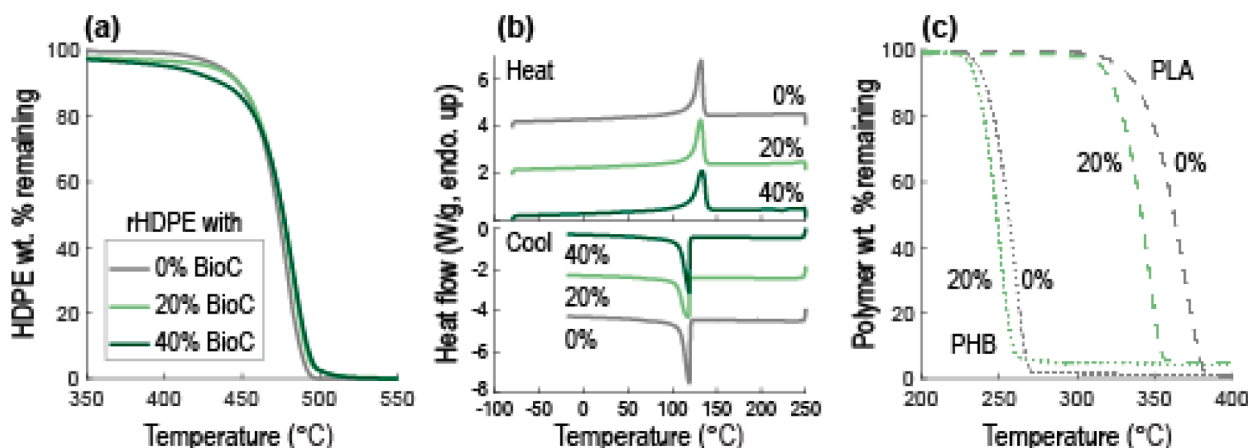


Fig. 2. Thermal characterization of rHDPE and biochar composites with 0%, 20%, and 40% biochar addition showing (a) TGA curves in the region of rHDPE degradation, (b) DSC heating (top) and cooling (bottom) curves, and (c) TGA curves for composites of PLA and PHB with 0% and 20% biochar addition, showing the decrease in thermal degradation temperature with the addition of biochar.

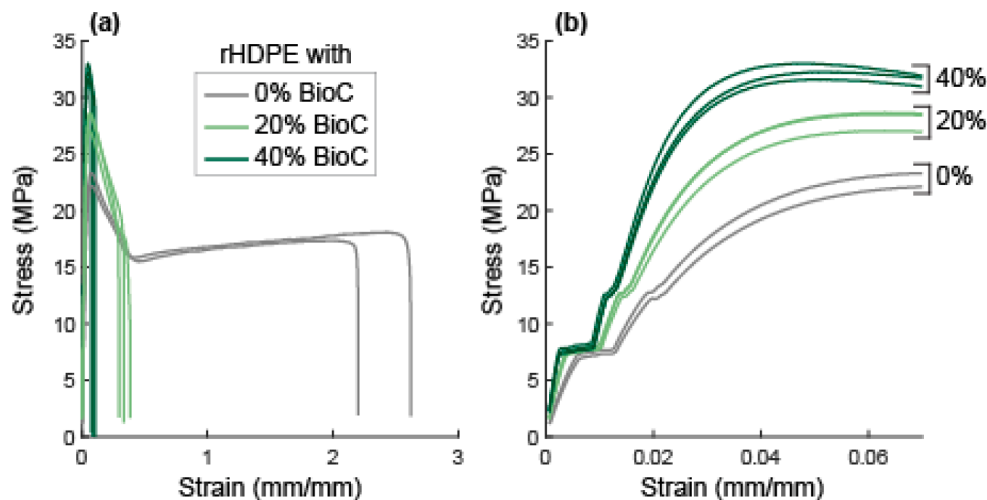


Fig. 3. Stress-strain curves from tensile test data of rHDPE with 0%, 20%, and 40% biochar addition, with (a) the full range of strain and (b) a detailed view of the initial loading region prior to peak stress.

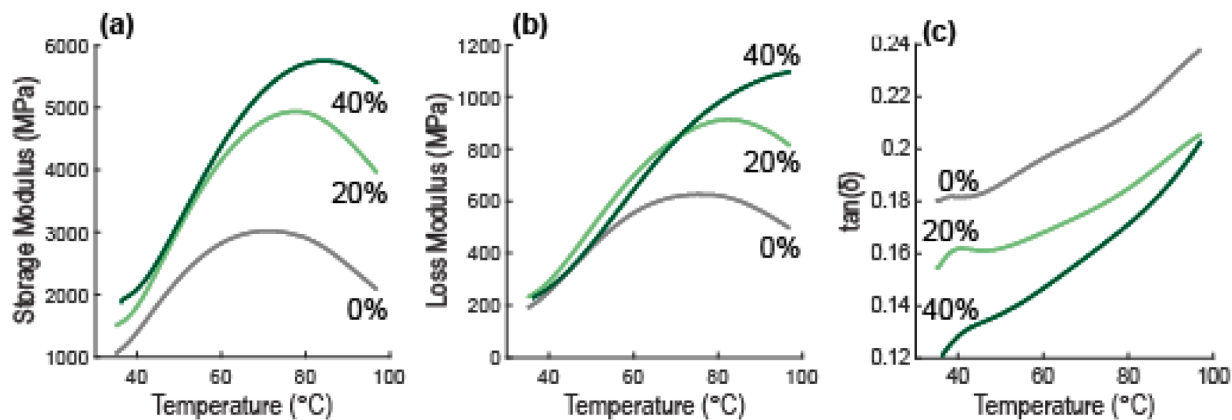


Fig. 4. DMA data for rHDPE composites with 0%, 20%, and 40% biochar, showing (a) storage modulus, (b) loss modulus and, (c) $\tan(\delta)$ as a function of temperature (see supplemental figure S2 for replicate curves).

energy than the production of virgin HDPE, PHB, or PLA. For rHDPE, the global warming potential and fossil fuel depletion values are driven by the energy required for injection molding and the negative global warming potential of the biochar. Differences in total impact between each plastic type, regardless of impact category, are primarily a result of differences in plastic production. The increased transportation-related impacts required to transport biochar for the addition were minimal relative to these differences between plastics.

4. Discussion

The increase in strength and stiffness of HDPE with the addition of biochar is consistent with the findings in past studies [11,12]. These studies have identified that the addition of biochar results in increased modulus and strength via three mechanisms: changes in polymer crystallinity, the high modulus of biochar, and polymer interlocking with the porous structure of biochar [16]. The large increase in crystallinity observed in this study with addition of biochar is consistent with these mechanisms and would be expected to result in a stronger and more brittle material as observed in this study [52,53]. As biochar has previously been found to have a high modulus relative to polymers [16], the increase in stiffness of rHDPE with the addition of biochar is jointly attributed to the increase in crystallinity of rHDPE and reinforcement by the biochar particles. The morphology of biochar composites was not examined in this study. However, past studies of wood-derived biochar

have found the highly macroporous structure of biochar to interlock with the polymer matrix, resulting in a good interface between the materials, which is expected to contribute to strong mechanical performance [15,16,34]. Further, past studies have found biochar addition to improve mechanical properties that were not examined in this study, including creep resistance, impact resistance, and flexural strength [12, 23]. While the impacts of biochar addition on virgin HDPE were not examined in this study, it is expected to result in similar shifts in mechanical behavior as in rHDPE.

A key finding of this study is that biochar could provide sufficient reduction in global warming potential to produce a cradle-to-gate carbon-neutral or negative material when added to plastic while increasing plastic strength. The amount of biochar needed to reach carbon neutral varies depending on the plastic type used. For example, the rHDPE examined in this study reaches 0 kg CO₂ equivalent with less than 40% biochar addition, while PLA and HDPE require 50% and 52% biochar addition, respectively. While the mechanical properties of composites containing higher biochar loadings were not examined in this study, past studies have shown similar strength and stiffness between 40% and 50% biochar addition in HDPE [12]. In contrast to the other plastics, PHB requires 72% biochar to reach 0 kg CO₂ equivalent global warming potential, which would be expected to result in a significant reduction in composite strength.

Forestry residues were examined as the feedstock for biochar in this study, and they were assumed to have no emissions for production

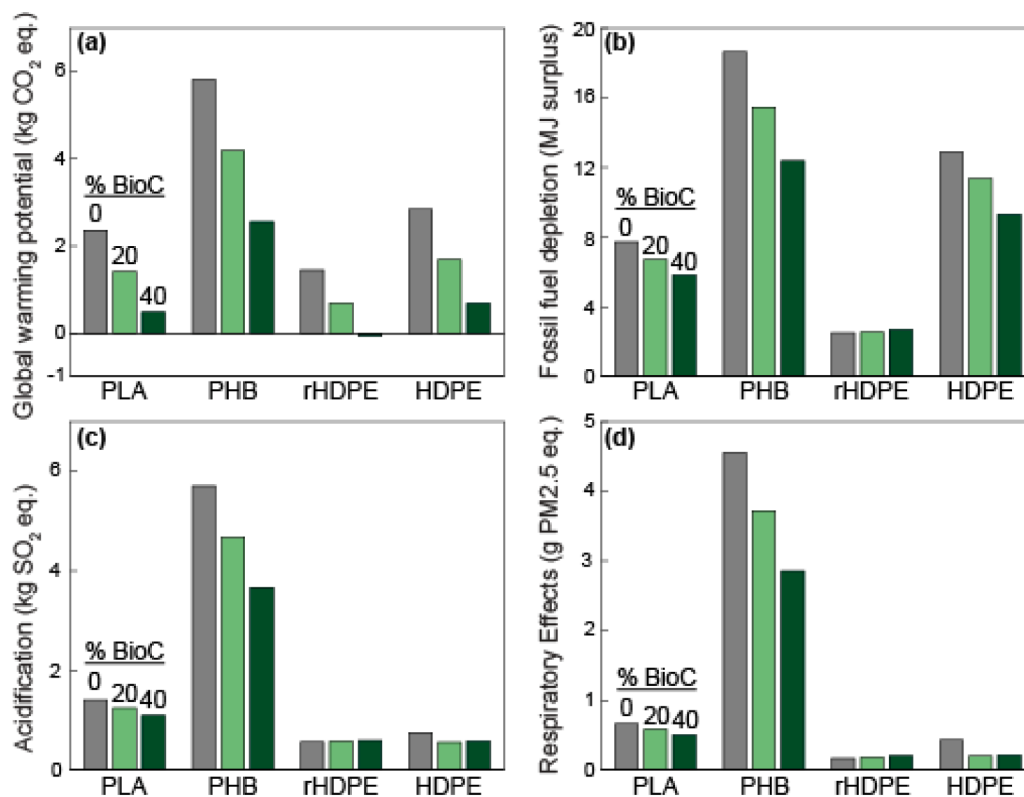


Fig. 5. Environmental impact assessment results for PLA, PHB, rHDPE, and HDPE for 0%, 20%, and 40% biochar addition showing impact indicators of (a) global warming potential, (b) fossil fuel depletion, (c) acidification, and (d) respiratory effects are shown. Other impact indicators in the TRACI weighting schemes are presented in supplemental figure S3.

(other than the uptake of CO₂ during photosynthesis that resulted in the carbon content of the biochar). However, many studies, including past life cycle assessment studies of biochar-filled composites, have examined the growth of biomass specifically to produce biochar [36–38]. These studies have identified crop cultivation as a critical area to reduce emissions in biochar production, which is avoided by using waste-derived feedstocks [6,38]. If biomass were cultivated for biochar production, additional emissions and land-use changes would need to be considered beyond what is shown in this work. Further, in this study, the avoided impacts of decomposition of the feedstock were not considered and may vary depending on how waste feedstocks would otherwise be disposed. This consideration may be more critical for high environmental impact waste feedstocks, such as food waste, where conversion to biochar may provide further reductions in global warming potential by avoiding the greenhouse gas emissions associated with the decomposition of the feedstock.

In addition to global warming potential considerations, shifts in material performance must be considered. The improvement in strength and stiffness of rHDPE with the addition of biochar and the brittle behavior at 40% biochar may change the target applications for the rHDPE-biochar composites. For example, the brittle behavior of 40% biochar is more similar to neat polyethylene terephthalate (PET) or polypropylene (PP) than neat HDPE, and high biochar content composites may be more desirable in applications where high failure strain is not needed [54]. In some applications, the impact on strength and stiffness from using biochar may allow for lower quantities of material to be used to meet the loading requirements, thus further reducing the environmental impacts of the final product.

If impacts are considered with a functional unit of volume per unit strength (e.g., m³/MPa), rather than mass as used in this study, rHDPE and biochar composites can achieve greater reductions in global warming potential. Namely, with a 20% biochar filler content, the

rHDPE composites achieve a 62% decrease in global warming potential on a strength basis relative to unfilled rHDPE, compared to a 48% decrease on a mass basis. Combined, this study's materials characterization and environmental impact assessment highlight how biochar may be applied to result in a stronger material, with lower environmental impact. In common applications of disposable plastic, such as food packaging or takeout containers, biochar could be applied as a filler to fulfill multiple roles – for example, acting as a colorant, increasing strength, increasing heat deflection temperature, while also reducing reliance on petroleum-derived materials and reducing total greenhouse gas emissions for the product. The improved properties of rHDPE with the addition of biochar could shift the polymer used in these applications, for example allowing for biochar-filled rHDPE to be used in takeout food packaging in place of PP, due to its higher stiffness and heat deflection temperature. If challenges with the thermal degradation temperature can be overcome, and similar mechanical increases to rHDPE achieved, this finding is particularly important for PLA and PHB, as more limited ranges of material properties are available for biodegradable plastics than petroleum-derived plastics [55].

The observed increase in strength with the addition of biochar is particularly important for rHDPE, as plastic recycling typically decreases the strength of the plastic [56]. The improved strength of rHDPE with the addition of biochar may expand the applications for which rHDPE has sufficient strength. Further, as the biochar examined in this study is currently inexpensive relative to rHDPE, biochar addition to plastic may improve the economic viability of the recycling industry [57,58]. Replacement of virgin HDPE or other plastics with rHDPE filled with biochar may further reduce the environmental impact of these materials as rHDPE had lower environmental impacts across most impact categories studied.

Despite these benefits to the mechanical properties of fossil-fuel-derived plastics, past studies have shown that the addition of biochar

to PLA and PHB reduces the thermal degradation temperature and molecular weight of these polymers, resulting in poor mechanical properties and processing challenges [22–28]. This decrease has been hypothesized to be due to the presence of inorganic compounds in the biochar catalyzing chain scission of the polymer [27,28]. As the Pacific Biochar examined in this study contained inorganics (Supplemental Figure S5), the mechanical properties of PLA and PHB were not studied herein. These inorganics may be removed with washing steps (e.g., with water or hydrofluoric acid) [59], but this processing step was not considered in our environmental impact assessment, as the exact processing depends on the biochar feedstock and desired level of inorganic removal. However, the addition of these processing steps would be expected to increase certain environmental impact categories (e.g., increased water use, additional energy required to heat water, or energy required to separate biochar after washing). Addressing this issue remains a key gap to the application of biochar as a filler material in PLA or PHB.

Importantly, as this study focused on the cradle-to-gate impacts of biochar addition to plastics, any changes to the material use phase or end-of-life impacts were not examined in this study. Depending on the desired application for biochar-filled plastic, it may be necessary to examine changes in material environmental wear and fatigue failure to determine the full life cycle impacts of biochar addition. Further, the biochar addition may impact the material's end-of-life depending on the disposal method. If the plastics are landfilled (rHDPE, HDPE, PLA, or PHB), composted (PLA or PHB), or degrade in the environment (PLA or PHB), the carbon in the biochar is expected to remain sequestered, and in the case of composted PLA or PHB, to positively benefit both the rate of composting [21,27] and the eventual compost application to

agriculture [5]. However, if the material is combusted, the carbon in the biochar will not remain sequestered, potentially increasing the overall greenhouse gas emissions of the material. Further, the addition of biochar to plastic currently makes the material challenging to recycle [60] and, therefore may reduce recycling rates for otherwise commonly recycled plastics, such as HDPE.

5. Conclusions

The environmental impact assessment conducted in this study highlights the potential for biochar to reduce certain environmental burdens, particularly the global warming potential and fossil fuel depletion, of a broad set of plastic materials. Further, our work suggests the addition of biochar improves the strength and stiffness of rHDPE, with a 45% increase in tensile strength and a 126% increase in tensile modulus at 40% biochar addition. A key finding of this study is that in rHDPE, HDPE, and PLA, biochar addition can result in a carbon-neutral material at the factory gate at 40–50 wt.% biochar addition. While use-phase and end-of-life stages were outside the scope of our environmental impact assessment, biochar addition to biodegradable plastics can increase biodegradation rates, while in landfilled plastics the biochar will continue to sequester carbon stored in biochar. This work supports the rapidly growing interest in applying biochar as a filler material in plastics by quantifying the potential environmental benefits of using biochar filler in a variety of plastics in addition to modifying the material properties.

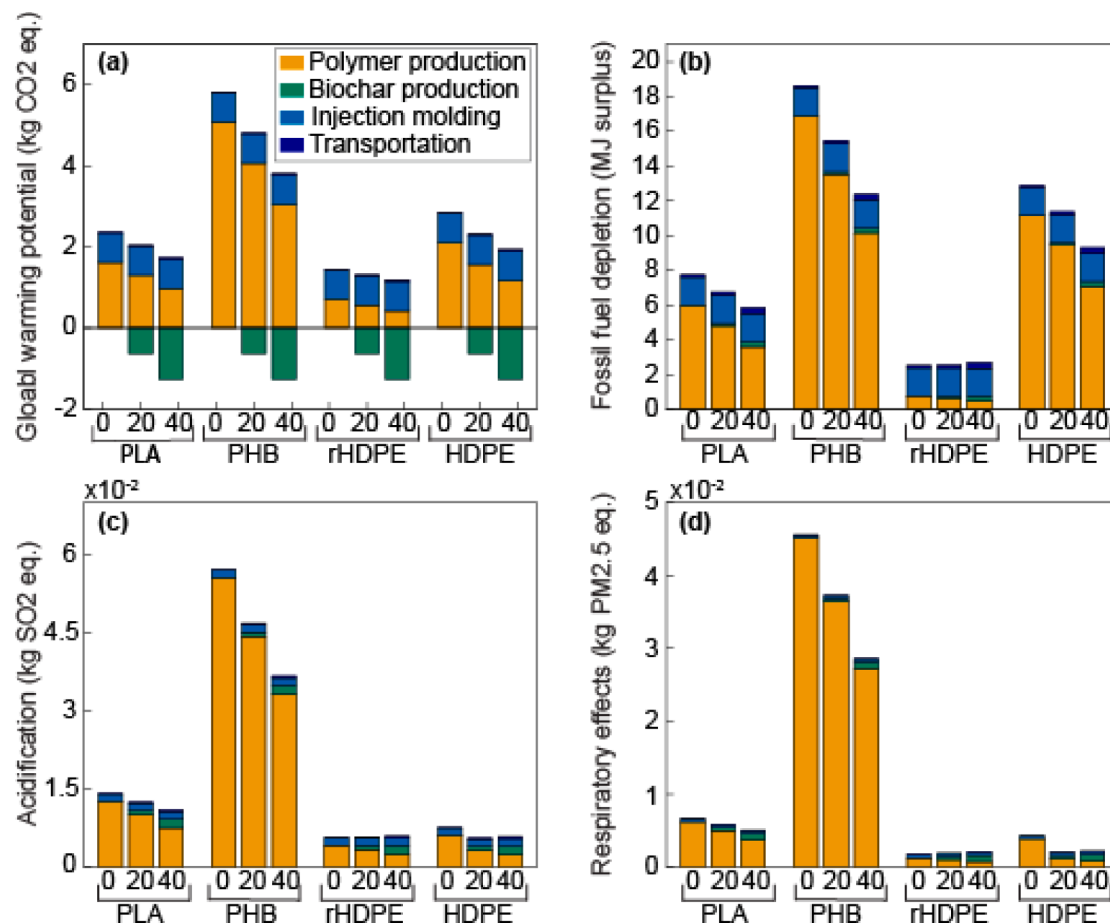


Fig. 6. Stacked bar plots of (a) global warming potential, (b) fossil fuel depletion, (c) acidification and, (d) respiratory effects by constituent and processing step for composites of 0%, 20%, and 40% biochar with HDPE, rHDPE, PLA and PHB. Other impact categories are shown in supplemental figure S4.

Declaration of Competing Interests

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

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.jcomc.2022.100253](https://doi.org/10.1016/j.jcomc.2022.100253).

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