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Ecological Engineering Research Group

# **Developing applications for biochar in a circular economy: a review of addition to biodegradable polymers**

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## **Abstract**

Increasing the application fields for biochar presents an opportunity to replace fossil-fuel derived products with carbon-neutral biomass. However, to do so successfully the biomass, process requirements, product-use and end-of-life must be appropriately researched and evaluated. In this review biochar applications beyond soil were conceptually mapped and the addition of biochar to produce composites was identified as a niche theme with biodegradable plastics in particular showing promising potential for further development in the context of a circular economy. A meta-analysis of 20 biocomposite characterisation studies found that biochar up to 20 wt % produced from various feedstocks and under differing pyrolysis conditions can lead to viable, or even biocomposites with improved properties for a number of bio-polymers. A set of recommendations for future biochar-PLA composite characterisations was suggested based on a qualitative evaluation of the publications under review. Due to the foundational nature of the research further biochar characterisation, biodegradation studies and sequential applications for biocomposites were identified as key future topics to evaluate if biochar-biocomposites are a promising application in a low-carbon circular economy.

# 1. Introduction

Biochar is a common name for a stable, carbon rich charcoal that is produced by the heating of biomass in the absence of oxygen (Burrell et al., 2016). The most commonly used processes for biochar production are gasification, torrefaction and pyrolysis (Meyer et al., 2011), which is the thermal decomposition of materials in an inert atmosphere producing syngas, liquid, or bio-oil and a solid product (char) (Kan et al., 2016). Various biomasses, including wastes can be utilised for biochar production. As the source of carbon in lignocellulosic biomass is atmospheric carbon dioxide and the produced char is recalcitrant the production and utilization of biochar has been recognised as a potential negative emission technology (NET) (Smith, 2016) by the Intergovernmental Panel on Climate Change (IPCC) and national bodies. It is promising, with a higher technology readiness level and lower cost than, for example, Direct Air Carbon Capture and Storage (McLaren, 2012). Whilst estimates for carbon savings are lower than other NETs it is considered attractive due to its low energy and water requirements, ability to utilise waste biomass and low cost of carbon transport and storage after pyrolysis (Smith, 2016).

The potential for carbon sequestration via biochar is largely linked to its historic application to soil (Lehmann, 2007) and how it functions as a soil amendment (Jeffery et al., 2011). If scaled, additional benefits such as bioenergy production, avoided emissions from biomass decay and soil sequestration can potentially offset 1.8 Gt CO<sub>2</sub>-C equivalent (CO<sub>2</sub>Ce) per year, 12% of current anthropogenic CO<sub>2</sub>-Ce emissions (Woolf et al., 2010). Whilst the climate-change mitigation potential is greater than combustion of the same biomass for bioenergy such scaling is likely to face a plethora of techno-economic limitations related to competition for biomass, legislative limitations surrounding contaminated feedstock use, availability of appropriate soil (McLaren, 2012) and relatively high cost for agricultural applications (Bach et al., 2016; Wurzer et al., 2019). Independently of these limitations biochar's physiochemical properties and capacity to be functionalised has also led to an expansion of research and early industrial applications in a wide variety of fields ranging from catalysts, fuel cells, filtration media (W.-J. Liu et al., 2015), addition to cementitious aggregates (Legan et al., 2022) and polymer composites (Chang et al., 2021). These are particularly promising as they open the possibility for utilising a wider range of feedstocks, can replace fossil fuel-derived substances with biochar, and produce higher value products. This replacement can generate additional carbon savings whilst embedding the carbon from the biochar in a material independent of or compatible with soil application at the end of life (EOL). This makes it potentially compatible with a circular economy approach to material production, use and disposal (Geissdoerfer et al., 2017).

Biochar's tuneable characteristics through pyrolysis and the potential to valorise waste feedstocks have led to its employment as a sustainable filler in common thermoplastics such as polyethylene and polypropylene (PE, PP) (Chang et al., 2021), wood composites (Das et al., 2016) and carbonaceous-reinforced thermosets (Bartoli et al., 2020). Whilst this addition can lead to improved material performance and emission reductions at the factory gate (Kane et al., 2022), the lack of research surrounding EOL of this more complex material contributes the production of waste whose treatment also presents new challenges (Narayan, 2011). The use of biopolymers provides an opportunity to integrate biochar in a more sustainable commercial plastic life cycles as part of a circular economy, in which virgin polymers are made from renewable or recycled raw materials. Compared with fossil-based plastics, bio-based plastics can have a lower carbon footprint and exhibit advantageous materials properties; moreover, some offer biodegradation as an EOL scenario if performed in controlled or predictable environments (Narayan, 2011; Rosenboom et al., 2022). Bioplastic production is however not without trade-offs, including negative agricultural impacts, competition with food production, unclear EOL management, higher costs and possibly poorer performance (Babu et al., 2013).

## 1.1. Research Aim

The aim of this literature review is to use bibliometric methods to investigate the conceptual structure of biochar applications beyond soil, particularly the integration of biochar in biodegradable plastics. By systematically reviewing publications for explored feedstocks, process parameters, desirable composite characteristics and consideration of EOL scenarios the potential for integration in a circular economy will be investigated.

## 2. Materials and Methods

A bibliometric analysis using the statistical computing language R (R Core Team, 2022) and the package *bibliometrix* (Aria & Cuccurullo, 2017) was performed to conceptually map non-soil uses for biochar and composite research. To ensure the bibliometric analysis was reproducible a set of queries (Table 1) were used to search for publications from the Web of Science Core Collection database (Clarivate) due to its standardised metadata and comparable coverage to other renowned databases such as SCOPUS. Each query was analysed for the co-occurrence of *Keywords Plus*, a set of keywords generated by an automatic computer algorithm based on words appearing frequently in the titles of an article's reference (J. Zhang et al., 2016). The co-occurrence was examined using network analysis, thematic mapping and multiple correspondence analysis to determine the conceptual structure of the topic.

Query	Rationale	Publications	Date range
TS = biochar NOT soil	Capture scope of all biochar research not related to soil applications.	11,253	1998-2022
TI = <i>biochar</i> OR <i>biocarbon</i> AND TI = <i>*polymer*</i> OR <i>*plastic</i> OR <i>*plastics</i>	Search terms used to isolate studies examining biochar in relation to polymers and plastics.	120	2011-2022

Table 1: Queries used to collect publications for bibliometric analysis of biochar applications beyond soil. TI is an abbreviation for Title and TS is an abbreviation for Topic (default search attribute in Web of Science).

An additional query (Table 2) was used to perform a more detailed meta-analysis on the addition of biochar to common biodegradable polymers. Of the 35 publications, 8 were removed as they did not include biochar addition to biodegradable polymers for continued material use.

Query	Publications	Date range
TI = biochar OR biocarbon AND TS = <i>polyhydroxy*</i> OR <i>polylactic acid</i> OR <i>PLA</i> OR <i>polybutylene</i> OR <i>PBAT</i> OR <i>PHBH</i> OR <i>PHBV</i> OR <i>thermoplastic starch</i> OR <i>PCL</i>	35	2015-2022

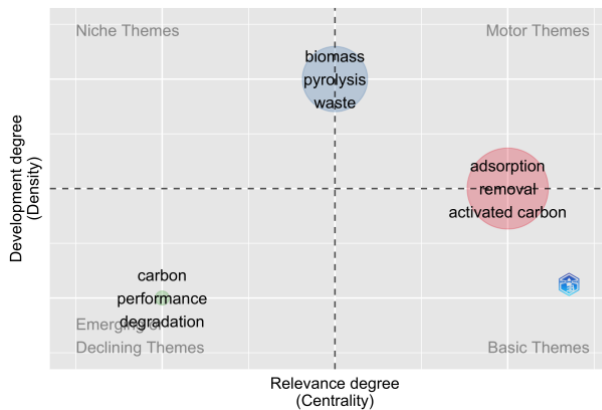
Table 2: Query used to collect publications related to biochar addition to biodegradable polymers. TI is an abbreviation for Title and TS is an abbreviation for Topic (default search attribute in Web of Science).

### 3. Results and Discussion

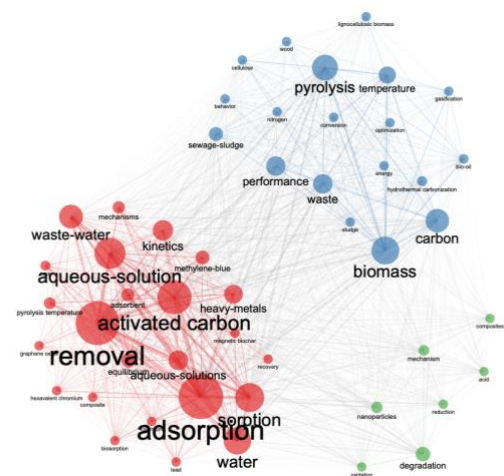
#### 3.1. Bibliometric Analysis

##### 3.1.1. Biochar beyond soil

Biochar and its applications beyond soil proved to be a well-researched field, with 11,253 publications ranging from 1998 and 2022 found through the query “TOPIC =biochar NOT soil”, which represented 49% of all literature on Web of Science related to biochar at the time (01.05.2022). A thematic analysis of *Keywords Plus* (Figure 1 (a)) indicates three clusters, each occupying distinct areas of the figure. Publications related to adsorption and pollutant removal (red), represented by “adsorption”, “removal” and “activated carbon” occur most frequently and show a high degree of centrality, implying strong links to other themes. Research regarding pyrolysis and feedstock selection (blue) exhibits decreased centrality and increased density, highlighting that the topics within this cluster are strongly linked within it and therefore strongly developed. Lastly publications related to “carbon”, “performance” and “degradation” are considered an emerging theme, with research within the topic less developed and not well-linked to other themes. By examining the underlying network in Figure 1 (b) additional relevant keywords such as composites identified as part of the emerging themes cluster (green). The keywords *biocomposites* or *circular economy* were not prevalent enough to appear in either figure.



(a) Thematic map of co-occurrence. The density axis refers to how close individual clusters are linked to one another whereas the centrality axis defines how interlinked the cluster is with other terms. Each quadrant represents a different theme, with bubble size drawn in proportion to the frequency of documents in which the keyword is used.



(b) Network analysis, the size of the individual nodes refers to the frequency of co-occurrence with other terms, with line width between nodes denoting the frequency of that specific co-occurrence.

Figure 1: Analysis of the co-occurrence of *Keywords Plus* in literature discovered using the query TOPIC = biochar NOT soil.

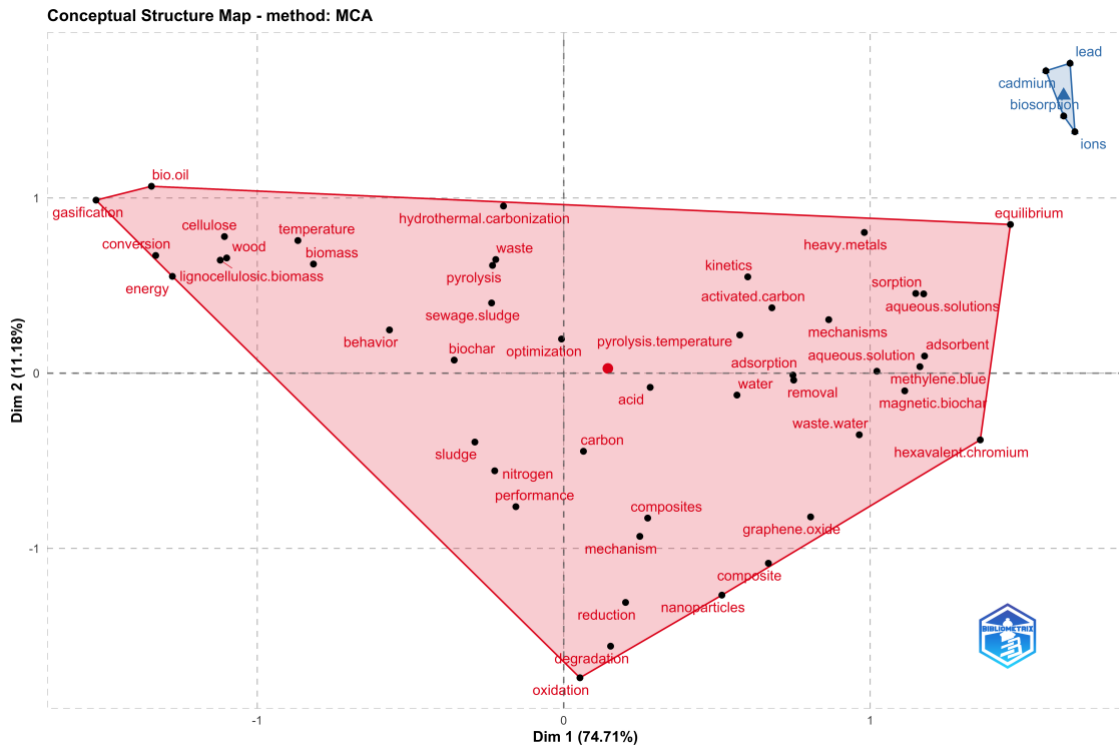


Figure 2: Multiple correspondence analysis of *Keywords Plus* using the query *TOPIC = biochar NOT soil*. The proximity of points represents how related they are in the specific dimension. Clusters, fields with close proximity, are denoted by differing colours.

The multiple correspondence analysis (Figure 2) shows that 85.89% of the variability in *Keywords Plus* can be accounted for by the first two dimensions. The first dimension accounts for 74.71% of variability, with publications arranged from left to right arranged by whether biomass conversion or adsorption dynamics in solution were investigated. The second dimension, which accounts for only 11.18% of the variation is less clear. Lower values are assigned to high-performance material applications, with catalyst functions (oxidation, reduction) placed centrally. Research related to nanoparticles, composites and graphene were also in close proximity, with a tendency towards the right side of the axis, related to adsorption dynamics in polluted solutions. Again, neither biocomposites nor circular economy occurred frequently enough to be mapped under this query.

### 3.1.2. Biochar in plastic composites

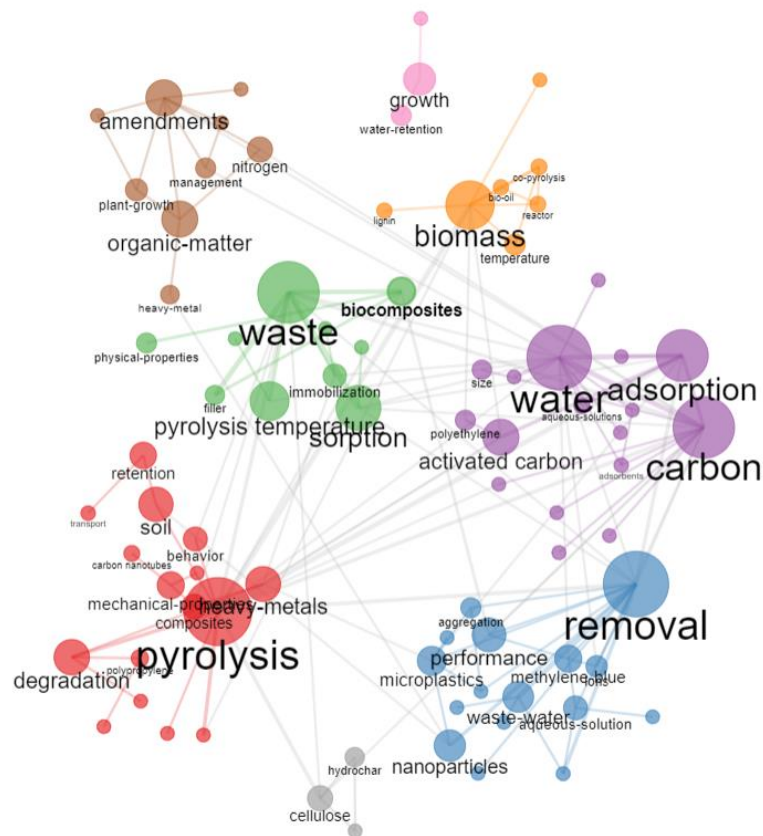


Figure 3: Network analysis of Keywords Plus related to plastics. Threshold for the number of keywords was increased to 200 due to low co-occurrence. Eight clusters can be observed with relatively poor number of connections.

Narrowing the query to focus on biochar integration in polymers reduced the number of results to 120, with a more recent publication date range of 2011-2022. When producing the thematic map of *Keywords Plus* several inconsistencies were observed, such as similar keyword labels (e.g., “heavy metal” and “heavy metals”) belonging to clusters with completely opposing density and centrality. Additionally multiple iterations of the script led to substantial variation of cluster positions across the diagram. When examining Figure 3 and cluster statistics a low number of keywords per cluster were identified, which likely led to the unpredictable results and highlights a limitation of the method. Despite the low number of keywords and interconnections Figure 3 shows eight distinct clusters. Whilst these could not be analysed for centrality or density the most frequently co-occurring node from each cluster summarised in Table 3. Biocomposites featured as a keyword linked to the “waste” cluster, with links to the keyword’s “performance”, “filler” and “physical properties”. This indicates that the production and characterisation of biocomposites containing biochar fillers from waste biomass is likely to be a core focus.

<b>Node</b>	<b>Cluster (color)</b>
pyrolysis	Red
removal	Blue
waste	Green
water/carbon	Purple
biomass	Orange
organic-matter	Brown
growth	Pink
cellulose	Grey

Table 3: Summary of most frequently used keywords in each distinct node

The multiple correspondence analysis (Figure 4) accounts for only 30.93% of variability, further highlighting that the query produced a set of poorly differentiated results. Interestingly, publications pertaining to co-pyrolysis and bio-oil were isolated via the first dimension in the blue cluster, highlighting that it is far removed



from most publications. Within the red cluster little clear thematic direction could be interpreted along the first dimension, other than soil-related keywords such as plant-growth, nitrogen, amendments, and organic matter are arranged on the left. A clearer transition was visible along the second dimension, with keywords largely related to pollutant removal on the bottom and a more diverse set of keywords related to physical properties of materials at the top. The key words composite and biocomposite being near keywords related to performance, mechanical properties and filler. This indicates that although the overall field of biochar addition to polymers varied substantially a separation between structural composite applications, energy use, soil applications and pollutant removal was observed. The lack of keywords related to sustainability or circular economy throughout highlights that these components, whilst undoubtedly mentioned are not central topics, with an insignificant number of publications directly exploring this aspect of biochar addition to plastic. The highly technical and performance-oriented keywords shows that publications found by the query are largely in the realm of foundational research.

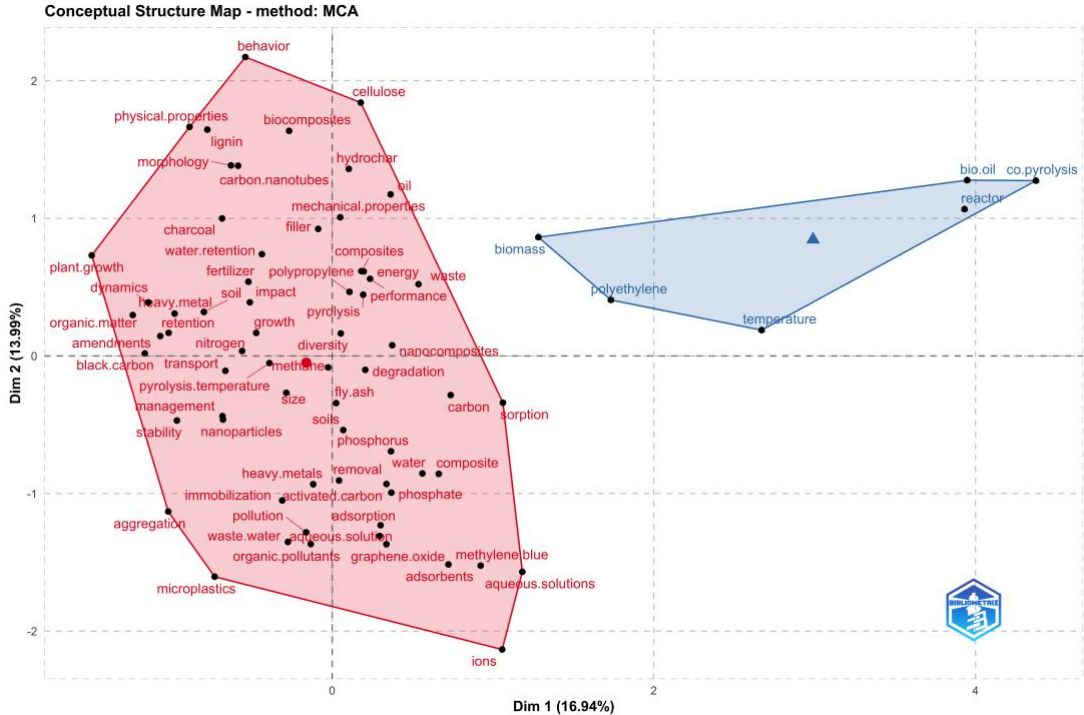


Figure 4: Multiple correspondence analysis of Keywords Plus of literature discovered when examining the combination of biochar and polymers. The proximity of points represents how related they are in the specific dimension. Clusters, fields with close proximity, are denoted by differing colours.

### 3.1.3. Limitations

Bibliometric approaches proved useful for mapping the field of research and can provide valuable insights into links between topics in a quantitative and reproducible manner. However, the methods for analysis require relatively large and well-developed topics in order to perform consistently. The choice of query and visualisation parameters (number of nodes) can also have substantial implications on the results and whilst reproducible, can be subject to bias.

## 3.2. Meta-analysis of biochar addition to biodegradable composites

Using a query to gather publications related to the combination of biochar and biodegradable polymers produced 27 publications after removing irrelevant entries (Table 2), published between 2015 and 2022. Of these 20 publications investigated the characteristics of composites produced with the addition of biochar, confirming the findings from Section 3.1.2. These were further examined in the meta-analysis. The remaining publications investigated how biochar and polymers influenced degradation of harmful compounds (Fan et al., 2022; Gonçalves et al., 2018; C. Liu et al., 2021; Rossi et al., 2022) as well as end uses such as biosensors (Sobhan, Muthukumarappan, Wei, Qiao, et al., 2021; Sobhan, Muthukumarappan, Wei, Zhou, et al., 2021) and advanced biocomposite fertilisers (Cen et al., 2021). These were not examined in the meta-analysis but used to provide additional contextual information where useful.

### 3.2.1. Biochar feedstocks, pyrolysis processes and characterisation

#### 3.2.1.1. Feedstocks

Category	Feedstock	Reference
Woody waste	Wood Chip/Waste	Musiol et al. (2022), Hernandez-Charpak et al. (2022), Pudelko et al. (2021)
	Bark (Pinus Sylvestris or birchwood)	Haeldermans et al. (2021b), Haeldermans et al. (2021a), Arrigo et al. (2020), Vidal et al. (2022)
Biomass from forest management	Beechwood	Botta et al. (2021), Zouari et al. (2022)
	Bamboo	Q. Zhang et al. (2020), (Q. Zhang et al., 2022)
Agricultural biomass production	Miscanthus	Nagarajan et al. (2016), Snowdon et al. (2019), Snowdon et al. (2017)
	Jatropha seed	Nyuk Khui et al. (2021), Law Nyuk Khui et al. (2021)
Organic residues from food processing	Spent coffee grounds	Diaz et al. (2020), Arrigo et al. (2020)
	Cassava rhizome, Durian peel, Corncob, Pineapple peel	Aup-Ngoen & Noipitak (2020)
	Chicken feather	Li et al. (2020)
	Pistachio shells	Haham et al. (2021)
Other biogenic waste	Anaerobically digested dairy manure	Hernandez-Charpak et al. (2022)
	Sewage sludge	Pudelko et al. (2021)
	Unspecified agricultural sludge	Haham et al. (2021)

Table 4: Summary of feedstocks used, grouped by approved carbon sink biomass categories (EBC, 2012)

Of the 16 distinct feedstocks examined woody biomass from waste or managed forests was the most prevalent category, featuring in 10 publications. Agricultural biomass from bioenergy crops such as *Jatropha* and *Miscanthus* grass were examined by 5 publications. Overall 7 different organic residues from food processing and 3 biogenic waste feedstocks were examined. Only three studies (Aup-Ngoen & Noipitak, 2020; Hernandez-Charpak et al., 2022; Pudelko et al., 2021) directly compared different biomass feedstocks for various polymer matrices and found that feedstock type had a substantial effect on mechanical properties (tensile strength) and thermal behaviour across all examined combinations. The physical target properties of biochar, specifically porosity, moisture, carbon content and functional groups were identified as key when choosing a compatible feedstock (Hernandez-Charpak et al., 2022). These properties largely influence the hydrophilicity of biochar relative to the hydrophobic polymers used, which defines the compatibility between the two and influences the properties of the final properties (Zouari et al., 2022).

### 3.2.1.2. Pyrolysis processes

Pyrolysis process	Temperature (°C)	Heating rate (°C/min)	Residence time (minutes)	Reference
Slow pyrolysis	450	NA	12	Haeldermans et al. (2021b)
	450	NA	NA	Diaz et al. (2020)
	550	NA	NA	Zouari et al. (2022)
				Pudełko et al. (2021)
	600	5	30	Q. Zhang et al. (2020)
	650	NA	NA	Musioł et al. (2022)
				Snowdon et al. (2019)
	450	NA	12	Haeldermans et al. (2021a)
	400, 500, 600	5	120	Aup-Ngoen & Noipitak (2020)
	300 & 600	NA	30	Li et al. (2020)
Microwave pyrolysis	500	10	120	Q. Zhang et al. (2022)
	700	5	60	Arrigo et al. (2020)
	180	NA	NA	Nyuk Khui et al. (2021) Law
				Nyuk Khui et al. (2021)

Table 5: Summary of recorded pyrolysis process parameters. NA indicate no further information was given or made available via a data sheet

Pyrolysis conditions were not recorded across all studies, with many utilising commercially produced biochar and not providing a data sheet. Of the 20 studies only 12 specified the pyrolysis process, which was predominantly slow pyrolysis (10) followed by microwave pyrolysis (2). A range of temperatures, from 300 to 700 °C were examined. Residence times also varied from as low as 12 minutes to 3 hours (Table 5). Only Aup-Ngoen & Noipitak (2020) and Li et al. (2020) examined the effect of changing pyrolysis temperature, finding that H/C ratio decreased with higher temperatures due to the removal of functional groups and loss of volatile matter. Li et al. (2020) determined in mechanical testing that 20 wt % biochar at 600 °C had improved tensile and flexural strength compared to biocomposites produced with biochar pyrolysed at 300 °C. This is in line with literature findings that greater pore development induced by the effusion of volatile matter and reduced hydrophilicity improves biocomposite mechanical properties Ho et al. (2015). Consequently both studies continued biocomposite characterisation with higher temperatures of 600 °C. Only Q. Zhang et al. (2020) examined additional processing steps such as activation with 25% H<sub>3</sub>PO<sub>4</sub>. The resulting biocomposites high surface area (312.17 m<sup>2</sup> per g) and lower hydrophilicity was attributed to the improved thermal and mechanical properties. Consequently, pyrolysis conditions can have a substantial effect on the quality of the final biocomposite and should be regarded as an opportunity to optimise biochar properties for specific biocomposite applications.

### 3.2.1.3. Characterisation

Biochar characterisation predominantly involved examining physical and structural properties, with the most common characteristics and methods utilised summarised in order of frequency in Table 6. Particle size was of particular importance, as it greatly influenced dispersion, with particles below 75 µm resulting in more homogeneous mixture and improved biocomposite properties (Nagarajan et al., 2016). This was followed by determining functional groups, as the hydrophilicity of lignocellulosic fillers negatively impacts composite performance due to poor compatibility with hydrophobic polymers. Some publications used H/C from CHNS analysis as a proxy of functional groups, with higher ratios indicating fewer functional groups and consequently lower hydrophilicity/increased hydrophobicity. Additional morphological properties, such as specific surface area also compound the hydrophilic properties of biomass and influence the properties of the final composites.

<b>Characteristic</b>	<b>Method</b>
Particle size (10)	Sieving or laser diffraction
Structural properties (6)	Fourier-transform infrared spectroscopy (FTIR) and/or Raman spectroscopy
Specific Surface Area (5)	BET (Brunauer, Emmett and Teller)
Morphology (5)	Scanning electron microscope (SEM)
Proximate analysis (5)	Appropriate ASTM Methodology
Phase identification (2)	X-ray powder diffraction (XRD)
CHNOS (2)	CHNS
Elemental analysis (2)	Energy dispersive spectroscopy (EDS)

*Table 6: Summary of methods used to characterise biochar, sorted by decreasing frequency.*

Six of the 20 studies utilised commercial biochar, whilst no data sheet was provided the assumption was made that the product is manufactured according to the required safety guidelines of the country of production. None of the remaining publications investigated the presence of potentially toxic elements or organic contaminants (PAHs, PCBs, benzopyrene, benzofluorane) which means none of the materials would be EBC-ConsumerMaterials or BasicMaterials certified (EBC, 2012). Furthermore, yield was often not provided, which limited any investigation of techno-economic feasibility or life cycle assessments.

### **3.2.2. Composite production**

#### **3.2.2.1. Polymers**

##### **Poly(lactic Acid (PLA))**

The most commonly researched biopolymer was polylactic acid (PLA). PLA is used for various applications such as biomedical parts, food packaging, textile fibres and 3D printing filament. The demand for PLA is continuously growing, however PLA has some limitation for different specific applications owing to PLA brittleness, poor heat resistance, low toughness and poor biodegradability (Murariu & Dubois, 2016). The addition of biochar generally leads to improvements in the stiffness of biocomposites at the cost of tensile strength. However, when compared to other polymers such PP and PCL it was found to be sensitive to moisture within biochar, resulting in reduced mechanical performance (Hernandez-Charpak et al., 2022).

##### **Polycaprolactone (PCL)**

PCL is an aliphatic polyester, used in biomedical fields, microelectronics, adhesives and packaging due to its easy processability, biocompatibility and biodegradability (Woodruff & Hutmacher, 2010). Further potential industrial applications of PCL are limited due to its poor mechanical properties, including low flexibility and softness. Consequently, it was the second most prevalent polymer used to examine the effect of biochar addition. Hernandez-Charpak et al. (2022) found a significant decrease in tensile strength, elongation at break and a significant increase in elastic modulus when 10 wt % biochar was added - with similar patterns observed for PLA. Diaz et al. (2020) also observed increased brittleness with biochar addition to 50:50 PCL and thermoplastic starch composites. Whilst PCL can be produced from sugars and is more easily degraded than PLA, it is currently derived from fossil fuels on an industrial scale and requires non-renewable carbon sources in the polymerisation phase. Vidal et al. (2022) found that oxidised and exfoliated biochar addition (up to 1 wt %) during polymerisation acted as a sustainable, multifunctional initiation site that promoted the growth of PCL. The final composite exhibited improved mechanical properties, crystallinity, thermal stability and degradation.

##### **Poly(3-hydroxybutyrate) (PHB)**

PHB-based composites were only investigated by Haeldermans et al. (2021a), as despite its better biodegradability it remains relatively limited due to its narrow processing window and high commercial costs. Biochar addition led to PHB losses during processing, increasing with higher biochar proportions. This was likely a result of the solid particles increased shear stress and produced temperature hotspots where thermal degradation of PHB occurred, combined with destabilisation of PHB due to inorganic oxides (CaO, MgO etc.) found on the surface of biochar. High additions of 50 wt % also contributed towards deterioration thermal stability of the produced biocomposite.

## Polybutylene adipate terephthalate (PBAT)

PBAT, a promising biopolymer for various films was only examined by Botta et al. (2021) to produce biocomposite blown films. The addition of up to 10 wt % biochar to PBAT led to an improvement of the elastic modulus and increased hydrophobicity without compromising the film blowing process. Increased photo-oxidative resistance of PBAT films containing biochar were considered as promising qualities for application as film for packaging, agricultural and composting bags.

## Compatibilisers and additional polymer mixes

A common practice used to improve composite properties is the addition of different polymers to act as compatibilisers. Thermoplastic starch (TPS) was identified as an excellent mediator between biochar and PHB (Haeldermans et al., 2021a) during melt-compounding and reduced thermal degradation with increasing biochar quantities. Diaz et al. (2020) also successfully utilised a 50:50 blend of TPS and PCL and various biochar additions in thermoforming processes. However no treatment without TPS was included, preventing comparisons. Maleic-anhydride-grafted-PLA was added by Li et al. (2020) with the goal of increasing adhesion between filler and matrix and led to improved impact strength in 20 wt % biochar when compared to the uncompatibilised sample (17.23 J/m compared to 14.32 J/m - neat PLA exhibited an impact resistance of 22.9 J/m). Several studies also examined the successful addition of non-biodegradable polymers such as polypropylene (PP) (Snowdon et al., 2017) and polytrimethylene terephthalate (PTT) (Nagarajan et al., 2016). However quantities of PP and PTT (> 50%) limit the comparability of these results to others within the subset. The addition of biodegradable compatibilisers can therefore be useful in allowing potentially unsuitable biochars to be used in biocomposites or further tuning the performance of successful biochar-biopolymer pairings.

## Addition amount

Biochar addition varied, with all publications settling on two distinct addition ranges. These were either between 0.1-7.5 wt % in increments < 1 wt % or a range between 10-50 wt % with larger increments of 10 or 20 wt %. A comparison of tensile strengths (Table 7) indicates that increased biochar addition led to decreasing tensile strength. This is attributed to the aggregation of high content biochar leading to micro-cracks, in line with the previously identified statements regarding the importance of small particle size and homogeneous dispersion for improved mechanical properties. Whilst strength requirements are usage dependent, it was clear that additions beyond 40 wt % generally led to widespread degradation of thermal and mechanical properties.

Biochar loading (wt %)	PLA tensile strength (MPa)	Composites tensile strength (MPa)	Reference
0.25	~57	~51	Aup-Ngoen & Noipitak (2020)
7.5	~54	~50	Arrigo et al. (2020)
20	~68	~35	Pudelko et al. (2021)
40	~59	~19	Q. Zhang et al. (2022)

Table 7: Relationship between tensile strength and biochar addition

### 3.2.2.2. Comparison to conventional fillers

The practice of adding fillers to alter composite properties is fairly commonplace and the addition of biochar is often justified as a functional and more sustainable option to conventional fillers. One frequently investigated conventional filler is the clay mineral talc, which improves mechanical composite properties at the cost of increased density and reduced biodegradability. Only Snowdon et al. (2019) directly compared biochar to conventional talc filler in a fully biodegradable polymer. Both fillers reduced abrasion in a comparable manner, with biochar-filled composites exhibiting superior flammability resistance. Biochar addition did however result in increased permeability, making it less suitable for barrier applications. Another popular filler that has the shared advantage of being biodegradable and low-density is micro-crystalline cellulose (MCC), which was compared to activated biochar by Q. Zhang et al. (2020). MCC added at 50 wt % led to reduced mechanical and thermal properties, which was improved by the addition of activated biochar, largely due to its reduced hydrophilicity compared to MCC. Q. Zhang et al. (2022) found that biocomposites containing biochar showed improved mechanical and thermal properties when aramid fibres and silica were added when compared to only biochar added at 40 wt %. The study however did not examine lower biochar additions and the addition of aramid reduces the biodegradability of the final composite. Biodegradable fibres from hemp were examined in by Zouari et al. (2022), where an addition of

5 wt % biochar to 30% hemp fiber composites improved tensile and flexural strength of the resultant biocomposite at the cost of reduced water repellence. Biochar is therefore an attractive filler for non-barrier applications due to its low density, superior flammability and abrasion resistance. Further functionalisation of composites with biodegradable fibers may also prove interesting in adding directional strength.

### **3.2.2.3. Biocomposite production process**

Composite production processes were largely governed by the desired properties of the final product. The vast majority of publications used micro-compounding or blending, followed by extrusion and injection molding at differing process parameters to produce the biocomposites. Two publications used hydraulic molding, stating the added benefit of being able to perform cold molding (Law Nyuk Khui et al., 2021; Nyuk Khui et al., 2021). Diaz et al. (2020) examined thermoforming to produce coffee cup lids, film blowing to produce films and foaming to produce foam products. Only Arrigo et al. (2020) compared differing methods, finding that melt mixing at high temperatures improved biochar dispersion at the cost of PLA degradation during the shearing process. Solvent casting at room temperature however led to composites with a higher tensile strength and modulus and sensitivity to increased biochar addition across rheological, mechanical and thermal properties. It is clear the biocomposite production must be aligned to the polymers used and purpose of the product, with micro-compounding, extrusion and injection molding proving suitable for generating comparable research.

### **3.2.3. Biodegradation**

Musiół et al. (2022) examined degradation in respirometer for 21 days and in abiotic conditions for 70 days with water at 70 °C. Biochar content had no influence on decomposition in laboratory composting conditions. Under abiotic conditions, all composites exhibited a similar degradation course as neat PBAT/PLA, with PLA degrading within 42 days and PBAT remaining largely unchanged after the same period. Increasing biochar above 15 % wt in the polymer reduced the decomposition due to entrapment, whereas amounts below 15 wt % encouraged degradation through the absorption of moisture across multiple studies (Musiół et al., 2022; Snowdon et al., 2017). Vidal et al. (2022) examined degradation of PCL films containing 0.1 wt % biochar, finding that degradation in aqueous solution without enzymes led to increased degradation (qualitative assessment via SEM). When examining the degradation of polymer films with lipase a complete degradation, with visible microbial activity was achieved within 28 days for films containing 0.1 wt % biochar. Whilst no control was provided this was considered promising as additives can also often have negative impacts on degradation of PCL composites (Fukushima et al., 2010). Whilst biochar shows degradation advantages in abiotic environments due to its hydrophilic nature, further degradation studies with varying biomass feedstocks and conditions (industrial composting, conventional composting, direct addition to soil) are required to better understand it's influence on biodegradation of biopolymers.

### **3.2.4. Recommended parameters for further biocomposite investigations**

The original intent of the meta-analysis was to utilise biocomposite performance data, combined with biochar characteristics to determine a recommended procedure for biochar production and addition to biocomposites, with the goal of using an accepted standard grade material to perform degradation, feasibility and carbon abatement studies. However, the relative novelty of the field, limited documentation of pyrolysis conditions, inconsistent biochar characterisations, varying standards used to determine composite performance and substantial number of variables means insufficient data is available to perform a reliable quantitative analysis. Nonetheless several key recommendations, based on a cross-examination of 20 biocomposite characterisations can be made to guide further work (Table 8).

<b>Process step</b>	<b>Recommendation</b>	<b>Justification</b>
Feedstock selection	Local waste biomass, preferably comparing multiple feedstocks	Feedstock choice should be governed by regional biomass contexts. Woody biomass was the dominant feedstock and commercially viable alternatives should therefore be explored. If resources do not allow, standard biochar or characterised material should be preferred.
Pyrolysis Process	300 and 600 °C with two set residence times of 15 and 30 minutes	Pyrolysis temperature and residence time affects the yield, carbon content, porosity and functional groups of the final biochar. These all have implications for the feasibility of the process and final biocomposite and comparisons need to be made to discover a satisfactory compromise.
Biochar processing	Milling to 20 - 75 µm	Improved dispersion in the polymer matrix led to better biocomposite properties across multiple studies.
Biochar characterisation	A full EBC characterisation according to EBC-ConsumerMaterials standards (EBC, 2012), particle size and FTIR	Biochar integration into biodegradable materials requires an understanding of potentially harmful components. Additional characterisation of biochar also opens the possibility of examining their influence on biocomposite properties and whether application to soil is permitted.
Addition amounts	5, 10, 15, 20 wt %	A substantial degradation of biocomposite properties beyond 20 wt %, regardless of underlying biochar characteristics was generally observed.
Polymer choice	PLA	Was the most commonly investigated biodegradable polymer with comparisons across literature possible.
Compatibiliser and other fillers	Addition of thermoplastic starch (up to 20 wt %) is recommended. Additional fibres should be biodegradable and appropriate for final use.	The addition of thermoplastic starch improved biocomposite properties without compromising the biodegradability. Biodegradable fibres can improve directional strength of the final biocomposite, however a compromise must be met to ensure brittleness is not too high.
Process	Micro-compounding, extrusion and injection moulding	Commercially viable process with a large body of literature to perform comparisons.

Table 8: Recommended process for further biochar biocomposite investigations

### 3.2.5. Future research required to develop application in a circular economy

The addition of biochar to create biodegradable composites is still in the foundational stage, with material and process optimisation currently a core focus. Consequently, the circular economy perspective is only examined when justifying the benefit of valorising waste biomass for biocomposite use or highlighting the biodegradability of the produced biocomposite. Clearly additional research is required in order to evaluate the full potential of biochar-biocomposites in a functioning circular economy.

#### 3.2.5.1. Characterisation of biochar

Despite biochar feedstock and pyrolysis conditions influencing composite characteristics a decoupling between biochar characteristics and its influence on biocomposite production was observed for most publications. Instead of utilising commercially produced biochar with poor documentation, the examination of extensively characterised standard biochar materials (Mašek et al., 2018) would drastically improve comparability and expand insights into how the material might interact with various biopolymers. Whilst exploring local and novel biomass for suitability is key, substantial added value can be generated by either utilising characterised biomass or performing the characterisations beforehand. These will allow for an improved understanding of production costs, carbon abatement potential, environmental impact and composite characteristics. A greater understanding of biochar feedstocks is also required to address appropriate end-of-life scenarios (Narayan, 2011). This is particularly relevant for contaminated waste streams, as specification standards for compostable plastics (ASTM D6400, D6868, ISO 17055, and EN 13432) require that the resultant compost should have no impact on plants and heavy metal content in the polymer material should be 50% lower than prescribed thresholds in the country of use.

#### 3.2.5.2. Sequential use

The sequential use of biochar and its products can lead to financial gains, whilst reducing the reliance on carbon from non-renewable sources (Wurzer et al., 2019). None of the examined publications investigated further utilisation of biocomposite beyond their initial use or the integration of biochar from other

applications. The field of biochar applications is rapidly developing, from functional materials such as adsorbents, catalysts and electrodes to integration in building materials. Consequently, increasing amounts of recalcitrant, biogenic carbon will be in use and provided material requirements are met (e.g., structural properties, potentially toxic elements) these could be viewed as viable feedstocks. Additionally, the application of produced biocomposites could be extended through the production of novel, slow-release fertilisers if loaded with nutrients prior to application (Cen et al., 2021; Daitx et al., 2020).

### **3.2.5.3. Biodegradation**

Considering that biodegradable polymers often have a limited useful product lifespan it is surprising that only three studies examine biodegradation of the produced biocomposite (Diaz et al., 2020; Musioł et al., 2022; Vidal et al., 2022). Moreover Diaz et al. (2020) did not examine the biodegradation of biocomposites containing biochar. Whilst no reason was provided, one explanation is that recalcitrant biochar would contribute to the final mass being relatively high. This highlights a key potential conflict of biochar addition to bioplastics, namely that compostable plastics standards require that 90%+ of the material carbon must be converted to CO<sub>2</sub> via microbial assimilation with 180 days or less. This is diametrically opposed to the carbon sequestration context that biochar is presented in (Schmidt et al., n.d.).

Nonetheless biodegradation and consequent soil applications is an important end-of-life scenario for biocomposites containing biochar. Beyond the significance of sequestering carbon via soil addition, biochar presence in composites generally lead to decreased degradation of biocomposites through moisture adsorption. Of the 27 papers found, three examined the positive effect of biochar addition to soil on the degradation of fungicides (Fan et al., 2022) and silver nanocomposites in soil (Gonçalves et al., 2018). Considering that biochar addition to compost can also lead to improved composting performance (Agegnehu et al., 2017) targeted studies isolating the degradation of polymers should be performed to determine whether biochar can offer an advantage in biocomposite composting.

### **3.2.5.4. Co-pyrolysis**

An alternative end-of-life treatment, particularly when biodegradability is limited or the required disposal conditions are not met, is co-pyrolysis. Considering the low thermal degradation resistance of biopolymers, low temperature pyrolysis or gasification can be used to recoup energy and useful chemicals produced from thermal decomposition of added polymers (Cornelissen et al., 2008; Saeung et al., 2021; S. Zhang et al., 2020).

### **3.2.5.5. Feasibility, life cycle assessment and carbon abatement potential**

Considering that cradle-to-gate analysis for biochar addition to conventional polymers has been performed (Kane et al., 2022), that methodology for determining the carbon footprint of bioplastics is established (Narayan, 2011) and guidelines on how to certify biochar for C-sink potential exist (Schmidt et al., n.d.) shows an important but resolvable gap in biochar-biocomposite research. This is largely due to the foundational nature of the research; however it is limited by the aforementioned lack of information regarding biochar feedstock, production and yield combined with limited exploration of biodegradation/end-of-life. Increasing the availability of this information will be critical in evaluating the techno-economic feasibility, environmental impact and carbon abatement of various biochar biocomposites. This is key to determine whether the application field can be integrated into a functioning low-carbon circular economy.



## 4. Conclusion

Biochar applications beyond direct application to soil covered a substantial amount of all research related to biochar and was driven by research related to adsorption and evaluation of the pyrolysis process for various waste feedstocks. The production of composites was identified as an emerging field, with a much smaller and more recent subset of publications identified, with insufficient publications to highlight well differentiated themes. The application of biochar to biodegradable plastics presented an even smaller, novel and foundational research field dominated by evaluating biocomposite characteristics. The meta-review of these characterisations indicates that biochar up to 20 wt % produced from various feedstocks and under differing pyrolysis conditions can lead to viable, or even biocomposites with improved properties for a number of bio-polymers. Due to the large number of influencing factors and inconsistent characterisations only a qualitative recommendation for future biochar-PLA biocomposites was suggested. Key topics such as biochar characterisation, economics behind feedstocks and their processing, biodegradation studies and potential for integration in sequential applications were identified as critical to determine whether biochar-biocomposites are a promising application for biochar in a low-carbon, circular economy.

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