




REVIEW PAPER

New trends in biochar pyrolysis and modification strategies: feedstock, pyrolysis conditions, sustainability concerns and implications for soil amendment

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Abstract

As a waste-derived soil amendment with a long history, biochar has received extensive attention for its capability to improve soil fertility/health; remove or immobilize contaminants in soil, water and air; and mitigate climate change. With the aim of producing engineered biochars with excellent performances, new trends in biochar pyrolytic production and modification strategies have emerged. This review critically summarizes novel pyrolysis methods (e.g., microwave-assisted pyrolysis, co-pyrolysis and wet pyrolysis) and modification approaches (e.g., mineral modification, photocatalytic modification, electrochemical modification) with a focus on (a) the mechanisms involved in environmental remediation processes including soil immobilization, contaminant adsorption and catalytic oxidation; (b) effects of feedstock and pyrolysis conditions on physicochemical properties; (c) sustainability considerations in novel modification and pyrolysis strategies; and (d) the feasibility of extrapolating the results from wastewater treatment to soil remediation. It is argued that in order to achieve the maximum net environmental benefits, 'greener' modification

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methods are warranted, and the risks associated with pyrolysis of contaminated feedstock in soil amendment and contaminant sorption can be minimized through various novel approaches (e.g., co-pyrolysis). Furthermore, novel pyrolysis methods can be combined with emerging modification strategies to synthesize more 'effective' biochars. Considering the similar aims of modification (e.g., increase surface area, introduce oxygen-containing functional groups, increase aromaticity), the applicability of several novel approaches could in future can be expanded from contaminant adsorption/degradation in aqueous media to soil remediation/fertility improvement.

KEYWORDS

clean water, engineered biochar, food security, green and sustainable remediation, soil pollution

1 | INTRODUCTION

Biochar has been used in environmental applications for millennia. Pre-Columbian Amazonians produced biochar by covering burning biomass with soil, thus forming a black soil (*terra preta de índio*) that can be used to increase soil fertility (Ahmad et al., 2014; Glaser, Lehmann, & Zech, 2002; Palansooriya et al., 2020). Biochar can be defined as 'a solid material obtained from the thermochemical conversion of biomass in an oxygen-limited environment' (IBI, 2015). Various preparation methods can be used to obtain biochar from a range of biomass feedstocks, such as pyrolysis, hydrothermal carbonization and gasification (Xie, Reddy, Wang, Yargicoglu, & Spokas, 2015), among which pyrolysis is the most widely adopted method owing to its relative simplicity of operation and operability at a range of scales. The resultant biochars can be used in a wide range of applications, including soil amendment and remediation (Beiyuan et al., 2020;

O'Connor et al., 2018; Shen et al., 2018), wastewater treatment (Palansooriya et al., 2019; Wu, Xia, et al., 2020; Zhang et al., 2020), flue gas treatment (Klasson, Boihem, Uchimiya, & Lima, 2014; Wang, Hou, et al., 2020) and climate change mitigation (Dissanayake et al., 2019; Igalavithana et al., 2019).

In order to synthesize engineered biochars with excellent performances, more studies have recently been conducted to investigate the effects of various modification strategies on biochar properties (Figure 1) (Sajjadi, Zubatiuk, Leszczynska, Leszczynski, & Chen, 2019; Wu et al., 2019; Yang, Zhang, Sun, Du, et al., 2019). Modification of biochar or pyrolysis conditions will affect both biochar physical properties (e.g., increase surface area and improve pore structure) and chemical properties (e.g., introduce certain functional groups and induce activated oxygen species on biochar surface) (Figure 1), promoting the adsorption or degradation of different contaminants depending on the specific approach

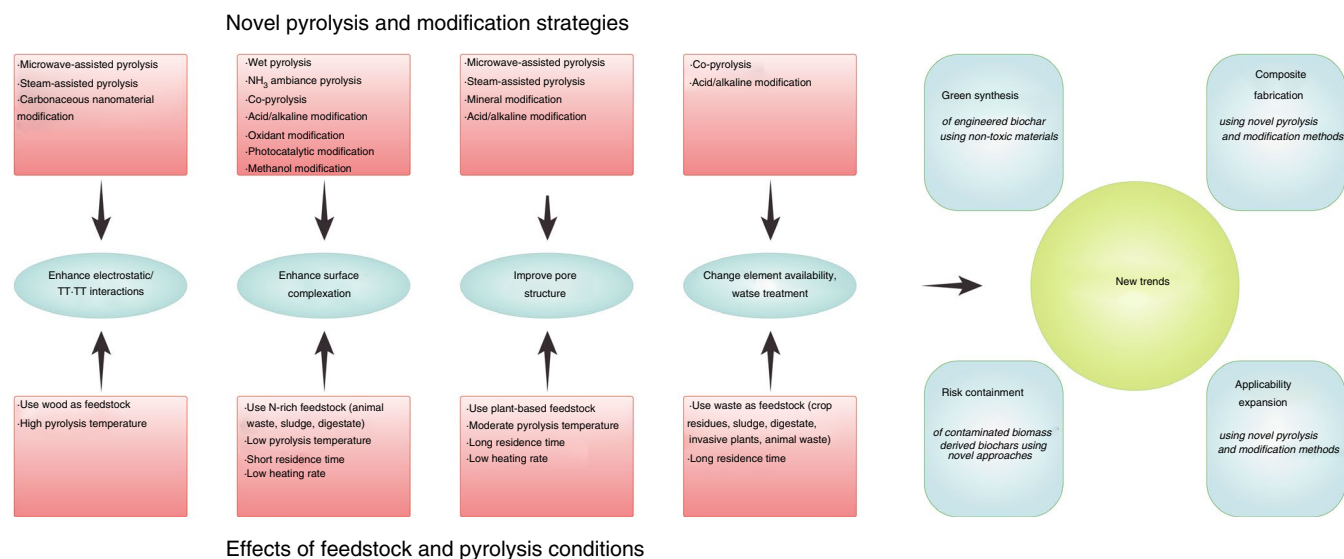


FIGURE 1 Major aims and new trends of novel pyrolysis and modification approaches with a focus on the effects of feedstock and pyrolysis conditions

employed (Ok, Chang, Gao, & Chung, 2015; Rajapaksha et al., 2016). In addition, with the emergence of the 'green and sustainable remediation' (GSR) movement (Hou & Al-Tabbaa, 2014; O'Connor, Hou, Ok, & Lanphear, 2020; You et al., 2017), more investigations have been conducted to produce 'greener' biochars, which are both environmentally and economically sustainable.

However, many novel attempts have only explored the contaminant sorption or degradation performances of engineered biochars in aqueous media to date. Considering the fact that many processes involved in contaminant immobilization and nutrient release take place in the aqueous phase, and that many mechanisms involved in contaminant adsorption and soil stabilization are quite similar (e.g., surface complexation, precipitation, ion exchange), discussion of biochar applications in wastewater treatment may provide fresh insights into soil remediation. The aims of this review are to (a) summarize recent advances in biochar pyrolytic production and modification, with a focus on the relationship between biochar properties and remediation mechanisms, (b) unveil new trends in biochar utilization in environmental applications with a focus on sustainability considerations and (c) investigate the feasibility of expanding the applications of some novel strategies into soil remediation/fertility improvement. The combination of novel strategies and the extension of biochar applicability to various fields are suggested, and several research directions are proposed.

2 | NEW TRENDS IN PYROLYSIS METHODS

Conventional pyrolysis methods such as slow pyrolysis and fast pyrolysis have been utilized for a long time to produce biochar, bio-oil and pyrolysis gas. However, in order to improve certain characteristics (e.g., specific surface area, oxygen-containing functional groups) of the resultant biochar or diminish the risks associated with the utilization of contaminated biomass in environmental applications, novel pyrolysis methods have emerged.

2.1 | Microwave-assisted pyrolysis

With wavelengths ranging from 0.001 to 1 m, microwave is a form of electromagnetic radiation that can be used as a dielectric heating method at certain frequencies (typically 2.45 GHz) (Li et al., 2016). Microwave radiation can transfer electromagnetic energy into heat without direct physical contact between the heat source and heated material, rendering a relatively high heating rate during the pyrolysis processes possible. Microwave heating mechanisms

involve ionic conduction, dipolar depolarization and interfacial polarization (Nam et al., 2018). During microwave-assisted pyrolysis processes, it is crucial that the feedstock absorbs microwaves effectively to cause polarization effects. It is believed that high water content favours the absorption of microwaves, since water can rotate and align in dipoles, resulting in collision and friction that account for the heating mechanism (Kong et al., 2019; Li et al., 2016). Microwave receptors such as activated carbon (Antunes, Jacob, Brodie, & Schneider, 2017) or metal-based microwave receptors (Li, Li, et al., 2019) have been added during pyrolysis to assure the absorption efficiency of microwave to help the system reach the desired heating rate and pyrolysis temperature.

Microwave radiation can affect both biochar morphology and chemical characteristics. Mašek et al. (2013) observed that microwave-assisted pyrolysis could achieve higher degrees of carbonization of biochar at lower pyrolysis temperatures compared with conventional slow pyrolysis, affecting the physical and chemical properties. Nair and Vinu (2016) found that narrow and deep pores (pore diameter 3.5 nm and pore volume $0.13 \text{ cm}^3 \text{ g}^{-1}$) were generated with the assistance of microwaves. Feedstock was heated homogeneously both inside and outside under microwave irradiation, and microscale explosions resulted in the distinct morphology. Omoriyekomwan, Tahmasebi, Zhang, and Yu (2017) observed that hollow carbon nanofibers were fabricated during pyrolysis of palm kernel shell. This is because the formation of an electric arc enhanced the self-extrusion of volatile matters from the inside the biomass and resolidification on the surface of resultant biochar. Paunovic et al. (2019) found that the number of oxygen-containing functional groups decreased through microwave irradiation, resulting in a higher adsorption capacity for naproxen because electrostatic interactions rather than surface complexation account for naproxen adsorption.

In environmental applications, biochars produced by microwave-assisted pyrolysis have been used for the adsorption of heavy metals, organic contaminants, phosphorus or nitrate (Table 1). This is probably because the decrease of oxygen-containing functional groups favoured both electrostatic interactions and π - π interactions between aromatic rings of biochar and contaminants (Paunovic et al., 2019; Zbair, Ahsaine, & Anfar, 2018). Several studies have also examined the metal adsorption performances of biochars pyrolysed with microwave assistance, but the results were not satisfactory, since surface complexation was suppressed (Elaigwu, Rocher, Kyriakou, & Greenway, 2014). It is therefore proposed that biochar produced through microwave-assisted pyrolysis may be effective for the remediation of soil contaminated with organic pollutants. Biochars produced during this process could also be used as bio-fertilizers for increasing the fertility and water holding capacity of sandy soils (Edeh, Mašek, & Buss, 2020; Mohamed, Ellis, Kim, Bi,

TABLE 1 Environmental applications of biochar pyrolysed with microwave assistance

Feedstock	Microwave	Environmental applications	Key findings	References
Dried sewage sludge (biosolids)	Heating power 600 W, activated carbon as microwave receptor	Adsorption of Ag	Ag adsorption capacity 70 mg g ⁻¹ , the resultant Ag/biochar nanocomposite could be used for methylene blue adsorption and photocatalytic degradation	Antunes et al. (2017)
Dried sewage sludge (biosolids)	Frequency 2.45 GHz, heating power 720 W	Adsorption of phosphorous	Adsorption capacity 147 mg g ⁻¹	Antunes, Jacob, Brodie, and Schneider (2018)
<i>Zea mays</i> (corn)	Residence time 18 min, frequency/heating power unknown	Increase pH of an acidic soil (liming effect, initial pH < 4.8)	Application of corn biochar increased soil pH substantially ($p < .0001$)	Chintala, Mollinedo, Schumacher, Malo, and Julson (2014)
<i>Prosopis africana</i> (wood)	Frequency 2.45 GHz, heating power 1600 W	Adsorption of Pb ²⁺ and Cd ²⁺	Adsorption capacity 45.3 and 38.3 mg g ⁻¹ for Pb ²⁺ and Cd ²⁺ , respectively	Elaigwu et al. (2014)
Oil palm shell	Frequency 2.45 GHz, heating power 900 W	Adsorption of methylene blue	Adsorption capacity 20 mg g ⁻¹ , surface area 410 m ² g ⁻¹	Kong et al. (2019)
Palm oil mill sludge	Heating power 500 W, irradiation time 25 min	Adsorption of trimethyltin, As and Zn	Zn adsorption capacity 44.5 mg g ⁻¹ , while trimethyltin and As adsorption was not satisfactory	Lam, Sum, Bashir, and Sethupathi (2017)
<i>Artemisia selengensis</i> (grass)	Frequency 2.45 GHz, with or without microwave receptor	Adsorption of methylene blue	Adsorption capacity increased with the addition of microwave receptor (ZnCl ₂ and Na ₂ CO ₃)	Li, Amin, et al. (2019)
Palm kernel shell	Heating power 700 W, irradiation time 25 min	Industrial effluent treatment	BOD reduction from 220 to 44 mg L ⁻¹ within 3 days	Liew et al. (2019)
Rice husk	Heating power 600 W, irradiation time 10 min	Immobilization of a Cd-contaminated river sediment (14.7 mg kg ⁻¹)	Microwave irradiation resulted in increased specific surface area and oxygen-containing functional groups, favouring the Cd stabilization	Liu et al. (2018)
Switchgrass	Frequency 2.45 GHz, heating power 750 W	Increase fertility and water holding capacity of sandy soil	Addition of K ₃ PO ₄ , bentonite or clinoptilolite as catalysts could increase surface area	Mohamed et al. (2016)
Switchgrass	Frequency 2.45 GHz, heating power 750 W	Improve plant growth, reduce bioavailability of Pb, Ni and Co in a sandy soil	Addition of K ₃ PO ₄ and clinoptilolite resulted in a high surface area of 405 m ² g ⁻¹ and high phosphorous content for metal precipitation	Mohamed, Ellis, Kim, and Bi (2017)
<i>Prosopis juliflora</i> (wood)	Frequency 2.45 GHz, heating power 600 W	Adsorption of Remazol Brilliant Blue R and methylene blue	Narrow and deep pores, surface area 357 m ² g ⁻¹ , adsorption capacity 83 mg g ⁻¹ and 91 mg g ⁻¹ for RBBR and MB, respectively	Nair and Vinu (2016)
Palm kernel shell	Frequency 2.45 GHz, heating power 550–750 W, irradiation time 30 min	Bio-fertilizer	Enhance the activity of living microorganisms (plant growth promoting bacteria), and increase nutrient availability	Nam et al. (2018)
Palm kernel shell	Frequency 2.45 GHz, heating power 2,000 W, activated carbon as microwave receptor	Synthesis nanofibre-shaped sorbent for metal adsorption	Electric arc generated during microwave-assisted pyrolysis resulted in nanofibre formation	Omoriyekomwan et al. (2017)

(Continues)

TABLE 1 (Continued)

Feedstock	Microwave	Environmental applications	Key findings	References
Plum kernel	Heating power 700 W, irradiation time 12 min	Adsorption of naproxen	Adsorption capacity 73.1 mg g ⁻¹ , mainly by electrostatic attraction	Paunovic et al. (2019)
Rice husk	Frequency 2.45 GHz, heating power 900 W, irradiation time 15 min	Tertiary treatment of wastewater	Adsorption capacity 71 and 497 mg kg ⁻¹ for phosphate and nitrate, respectively	Shukla, Sahoo, and Remya (2019)
Almond shell	Heating power 600 W, irradiation time 12 min	Adsorption of sulphamethoxazole	Adsorption capacity 344.8 mg g ⁻¹ , surface area 1274 m ² g ⁻¹	Zbair et al. (2018)
Sewage sludge	Heating power 1200 W, irradiation time 10 min, activated carbon as microwave receptor	Adsorption of eosin and safranin T	Adsorption capacity 43.66 and 70.78 mg g ⁻¹ for eosin and safranin T, respectively	Zhang, Tian, Yin, Zhang, and Drewes (2018)

Abbreviations: BOD: biochemical oxygen demand

RBBR: remazol brilliant blue R

MB: methylene blue

& Emam, 2016). Activities of plant growth promoting bacteria (PGPBs) could also be stimulated, resulting in better plant growth and crop yield (Nam et al., 2018).

2.2 | Steam-assisted pyrolysis

Several researchers have investigated the effects of steam during pyrolysis (as opposed to postpyrolysis steam activation) on biochar physicochemical properties (Table 2). Increased specific surface area and pore volume were observed, which were because of the removal of tar and other by-products on biochar surfaces (Krerkkaiwan & Fukuda, 2019; Lam et al., 2019). In this way, steam-assisted pyrolysis can promote the adsorption performance of biochars (Rajapaksha et al., 2014). It is noteworthy that apart from metal and organic contaminant adsorption from water, biochar produced by steam-assisted pyrolysis can also be used for sulphur dioxide (SO₂) removal from flue gas. Braghiroli, Bouafif, and Koubaa (2019) observed a decrease in oxygen-containing functional groups on the surface of biochar made from wood during steam activation. This resulted in increased aromaticity and intensity of delocalized π bonds (section 4.1.3) (Lewis base), thus favouring the adsorption of SO₂ (Lewis acid). Steam-assisted pyrolysis is easy to apply and is relatively energy-efficient compared with other methods aimed at increasing surface area (e.g., elevating pyrolysis temperature). As shown in Table 2, this pyrolysis method is being actively researched to improve the adsorption performances of biochars in aqueous media. It is suggested that the applicability of this 'green' method should be expanded (e.g., to soil remediation), since the mechanisms involved in adsorption of contaminants from the aqueous phase and in contaminant immobilization in soil are similar (e.g., physisorption, electrostatic attraction). It is hypothesized that biochars produced through steam-assisted pyrolysis could effectively immobilize organic aromatic contaminants such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs), since a decrease of oxygen-containing functional groups may result in enhanced π - π interactions, as has been described above.

2.3 | Wet pyrolysis

Wet pyrolysis is a novel method to introduce oxygen-containing functional groups such as hydroxyl and carboxyl onto the surface of biochar. Conversion of biomass into biochar and biochar modification can be achieved simultaneously in a wet pyrolysis system. Zhou, Wang, Huang et al. (2019), Zhou, Wang, Yao et al. (2019) mixed the biomass with phosphoric acid (85 wt.%) and stirred the mixture for 2 hr. After soaking, the mixture was poured into an autoclave and heated at 200°C for 5 hr. The resulting biochar

TABLE 2 Effects of steam on biochar properties and environmental applications of biochar prepared by steam-assisted pyrolysis

Feedstock	Pyrolysis temperature	Specific surface area ($\text{m}^2 \text{g}^{-1}$)	Pore diameter (nm)	Total pore volume ($\text{cm}^3 \text{g}^{-1}$)	Environmental applications	Performances	References
Black spruce and white birch	Fast pyrolysis at 454°C, followed by steam (0.3 L min^{-1}) activation at 900°C for 67 min	590	0.80	0.34	Adsorption of SO_2 from flue gas	Adsorption capacity 76.9 mg g^{-1}	Braghiroli et al. (2019)
Rice straw	Pyrolysis temperature 800°C, steam (100 ml min^{-1} , 30 min)	341	2.52	0.215	Catalytic decomposition of naphthalene	Conversion rate 76.9% (initial feed rate 0.013 g min^{-1})	Krekkaivan and Fukuda (2019)
Canola straw	Pyrolysis temperature 700°C, steam (5 ml min^{-1} as deionized water) for 1 hr	106	–	–	Adsorption of Pb(II) from wastewater	Adsorption capacity 195 mg g^{-1}	Kwak et al. (2019)
Soybean straw	Fast pyrolysis, steam (3 ml min^{-1}) at 800°C for 45 min	793	0.98	0.344 ^a	Adsorption of Cu^{2+} , Cd^{2+} , Ni^{2+} , Zn^{2+}	Adsorption capacity 95.7, 21.0, 30.0, 27.8 mg g^{-1} for Cu^{2+} , Cd^{2+} , Ni^{2+} , Zn^{2+} , respectively	Lima, Boateng, and Klasson (2010)
Pine sawdust	Pyrolysis temperature 550°C, steam (5 ml min^{-1} for 45 min)	397.1	–	–	Reduce greenhouse gas emissions in forest and grassland soils	Decreased microbial and enzyme activities accounted for the reduction in N_2O and CO_2 emissions	Pokharel, Kwak, Ok, and Chang (2018)
Tea waste	Pyrolysis temperature 700°C, steam (5 ml min^{-1} for 45 min)	576.09	2.00	0.109	Adsorption of sulphamethazine	Adsorption capacity 33.81 mg g^{-1}	Rajapaksha et al. (2014)
Invasive plant <i>Sicyos angulatus</i> L.	Pyrolysis temperature 700°C, steam (5 ml min^{-1} for 45 min)	7.10	8.39	0.038	Adsorption of sulphamethazine	Adsorption capacity 37.7 mg g^{-1}	Rajapaksha et al. (2015)
Mushroom	Pyrolysis temperature 800°C, steam (2 ml min^{-1} for 2 hr)	332	3.50	0.29	Adsorption of crystal violet	Adsorption capacity 1057 mg g^{-1}	Sewu, Jung, Kim, Lee, and Woo (2019)
Bamboo	Pyrolysis temperature 500°C, steam (5 ml min^{-1} for 45 min)	2.12	4.06	0.002	Adsorption of Cu^{2+} and tetracycline	Adsorption capacity 5.03 and 0.22 mmol g^{-1} for Cu^{2+} and tetracycline, respectively	Wang, Huang et al. (2019)

^a Micropore volume. Abbreviations: CEC: cation exchange capacity FTIR: fourier-transform infrared spectroscopy LDH: layered double hydroxide

TABLE 3 Environmental applications of co-pyrolysed biochar

Feedstock (weight ratio)	Pyrolysis conditions	Aims of co-pyrolysis	Environmental applications	References
Sewage sludge + manganese ore (20:1)	Pyrolysis temperature 450°C	Not mentioned	Reduce nutrient leaching from the soil (especially nitrates)	Duan et al. (2020)
Sewage sludge + waste tea (1:1)	Pyrolysis temperature 300°C, residence time 2 hr	Improve pore structure	Adsorption of Cd (17 mg g ⁻¹)	Fan et al. (2017)
Rape straw + phosphate rock (5:1)	Pyrolysis temperature 500°C, residence time 2 hr	Precipitate Pb (form pyromorphite)	Pb removal from water	Gao et al. (2018)
Rice husk + polyethylene (1:4)	Pyrolysis temperature 390°C, residence time 35 min	Not mentioned	Adsorption of Cr (III) (21.1 mg g ⁻¹)	Godinho et al. (2017)
Sewage sludge + bamboo sawdust (1:1)	Pyrolysis temperature 600°C, residence time 1 hr	Reduce metal mobility	Reduce the risks of metal leaching	Jin et al. (2017)
Rice husk + Mg/Al-LDH	Pyrolysis temperature 700°C, residence time 2 hr	Increase binding affinity of anionic contaminants of biochar	Adsorption of phosphate (70.8 mg g ⁻¹)	Lee et al. (2019)
Poultry litter + phosphate/MgO (2:1, P/Mg 1:1 molar ratio)	Pyrolysis temperature 500°C, residence time 2 hr	Improve soil fertility	Slow-release fertilizer for tropical soils	Lustosa, Penido, Castro, Silva, and Melo (2017)
Swine manure + rice straw (1:3)	Pyrolysis temperature 400°C, residence time 2 hr	Safe disposal of metal-containing swine manure	Soil metal (Pb, Cu, Zn, Cd) immobilization (dosage 3%)	Meng, Tao, Wang, Liu, and Xu (2018)
Sewage sludge + Fe/Ti-loaded chitosan	Pyrolysis temperature 800°C, residence time 1 hr	Sewage sludge disposal	Catalytic degradation of methylene blue	Mian and Liu (2019)
Rice straw + polyethylene terephthalate (4:1)	Pyrolysis temperature 550°C, residence time 4 hr	Increase hydrophobicity, electrostatic interactions	Adsorption of DNT (5.1 mg g ⁻¹), DCP (11.5 mg g ⁻¹), Pb (115 mg g ⁻¹), CrO ₄ ²⁻ (1.2 mg g ⁻¹), SeO ₄ ²⁻ (12.0 mg g ⁻¹)	Oh and Seo (2019)
Wood + polyvinyl chloride (3:1)	Pyrolysis temperature 700°C, residence time 10 min	Introduce Cl on biochar surface	Flue gas Hg(0) removal	Xu et al. (2016)
Swine manure + corn straw (1:3)	Pyrolysis temperature 700°C, residence time 2 hr	Reduce metal mobility (Cd, Pb, Ni, Cr, Cu, Zn)	Reduce the risks of metal leaching	Xu et al. (2019)
Sewage sludge + walnut shell (3:1)	Pyrolysis temperature 600°C, residence time 3 hr	Improve pore structure	Adsorption of ammonium (22.85 mg g ⁻¹) and phosphate (303.49 mg g ⁻¹)	Yin, Liu, and Ren (2019)
Sewage sludge + hazelnut shell (7:3)	Pyrolysis temperature 850°C, residence time 45 min, 4 M K ₂ CO ₃ activation	Improve pore structure	Adsorption of Cu(II)	Zhao et al. (2018)

Abbreviations: DNT: 2,4-dinitrotoluene DCP: 2,4-dichlorophenol LDH: layered double hydroxide

was mixed with sodium carbonate (to neutralize the system and promote the precipitation of heavy metals) and rinsed several times. A large number of phosphoric and oxygenic groups were present on the biochar surface, resulting in a high adsorption capacity for lead (Pb) (316 mg g⁻¹) and cadmium (Cd) (197 mg g⁻¹). Compared with conventional

pyrolysis procedures, wet pyrolysis is an energy-saving and cost-effective approach (Zhou, Wang, Huang et al. (2019), Zhou, Wang, Yao et al. (2019)). This method may become a new trend, as there is no need to create an oxygen-limited environment, making the pyrolysis conditions much simpler.

2.4 | NH₃ ambience pyrolysis

During ammonia (NH₃) ambience pyrolysis processes, NH₃ may react with oxygen-containing species (e.g., ketones, aldehydes, esters, furans) of biomass to form nitrogen (N)-containing heterocyclic compounds (i.e., pyrrole, pyridine, piperidine, indole) (Chen, Li et al., 2018). N-doped biochar can be utilized for metal adsorption. For instance, Mian et al. (2018) synthesized N-doped magnetic agar biochar through one-step heating of FeCl₃-laden biomass in the presence of NH₃. The resulting biochar could adsorb Cr(VI) effectively (adsorption capacity 142.86 mg g⁻¹), which was because of the surface complexation of Cr(VI) with N-containing groups and subsequent Cr(III) formation after the catalytic reduction of Cr(VI) by magnetic biochar. In addition, N-doped biochar could also promote the catalytic degradation process of methylene blue. Mian et al. (2019) found that NH₃ ambience pyrolysis could significantly reduce the bandgap energy of TiO₂/Fe₃O₄/biochar composite, making it possible for visible light with relatively low energy to excite electrons from the valence band to the conduction band. However, the mechanisms involved in this process were not fully understood. It is suggested that future research should investigate the role of N-containing groups in catalytic removal processes of contaminants in more depth, and examine the potential of N-doped biochars as a metal immobilizing amendment.

2.5 | Co-pyrolysis

Co-pyrolysis is an effective means to modify biochar properties and to synthesize composite products. In addition, it can be an effective way to reduce the environmental risks that may be associated with biochars produced from metal-rich feedstocks (i.e., sewage sludge, section 4.1.1) because of improved immobilization of inherent metals within the biochar matrix. After pyrolysis with unpolluted biomass (e.g., waste tea, bamboo, walnut shell, hazelnut shell), mobility of metals in the biochar could be reduced significantly, thus reducing the risks associated with metal leaching (Table 3). Similar effects have been observed by Buss, Jansson, and Mašek (2019), Buss, Jansson, Wurzer, and Mašek (2019) in the co-pyrolysis of woody biomass with biomass ash, showing a significant potential to reduce the availability of heavy metals by incorporation of the ash in biochar, while maintaining nutrients, such as potassium in their available forms (Buss, et al. (2019); Buss et al. (2019)). In addition, a synergistic effect on biochar carbon retention was observed. Co-pyrolysis can also enhance biochar functionality in environmental applications. Several studies found that co-pyrolysis could improve biochar pore structure and increase surface area (Fan et al., 2017; Zhao, Xu, Zeng, Li, & Chen, 2018). Apart from co-pyrolysis of biomass feedstocks, co-pyrolysis with plastics is

also a promising method for waste management, and the resulting biochars have been shown to adsorb contaminants effectively through electrostatic interactions (Oh & Seo, 2019). It is noteworthy that co-pyrolysis can be used to synthesize novel biochar composites. Lee et al. (2019) synthesized a biochar/layered double hydroxides (LDH) composite through co-pyrolysis after pre-loading MgAl-LDHs on the surface of rice husk powder through precipitation. Adsorption of phosphate was significantly enhanced, as anionic contaminants were effectively adsorbed by LDHs through ion exchange with negatively charged groups located between hydroxide layers.

3 | NEW TRENDS IN MODIFICATION STRATEGIES

There are many classes of novel modification strategies, with only the key ones reviewed in this work. It is noteworthy that the applicability of conventional modification methods (e.g., magnetic modification and acid/alkaline modification) has been expanded (i.e., from contaminant adsorption in wastewater to heavy metal stabilization in soil).

3.1 | Magnetic modification

Magnetic biochars have been widely researched as a sorbent for wastewater treatment. After modification with ferromagnetic elements (e.g., Fe, Co and Ni) and their oxides, biochars can be recycled easily through an external magnetic field, making the cleaning and regeneration processes much easier. A number of studies have examined the adsorption mechanisms and reusability of magnetic biochars (Chen, Ho, et al., 2018; Cho, Yoon, Kwon, Biswas, & Song, 2017; Karunanayake et al., 2017; Mian et al., 2018; Mohan, Kumar, Sarswat, Alexandre-Franco, & Pittman, 2014). Synthesis methods, adsorption performances and reusability of magnetic biochars have been reviewed elsewhere (Thines, Abdullah, Mubarak, & Ruthiraan, 2017; Wang & Wang, 2019; Wang, Zhao, et al., 2019).

However, these papers focused merely on the contaminant adsorption applications of magnetic biochars, while more recent studies have expanded the applications of magnetic biochars (Chen et al., 2019; Fu, Ma, Zhao, Xu, & Zhan, 2019; Li, Jiang, et al., 2019; Li, Lai, et al., 2019; Yi, Tu, Zhao, Tsang, & Fang, 2019). Owing to the capability to generate reactive oxygen species such as hydrogen peroxide (H₂O₂), sulphate radical (SO₄^{-•}) and hydroxyl radical (•OH), magnetic biochar can be used for the catalytic degradation of organic contaminants. For instance, Li, Lai, et al. (2019) modified pine needle-derived biochar with Fe-Mn binary oxides and examined its ability for naphthalene degradation

TABLE 4 Recent advances in mineral modification of biochars and their performances in environmental applications

Mineral	Aims of mineral modification	Environmental applications	Key findings	References
Attapulgite	Promote ion exchange	Adsorption of oxytetracycline adsorption (33.31 mg g ⁻¹)	Hydrogen bonding, ion exchange and complexation led to high adsorption capacity	Wang, Yang et al. (2019)
Attapulgite	Promote ion exchange, biochar act as supporting matrix	Immobilization of As and Cd in river sediment	Compared with virgin biochar, the clay–biochar composite had higher surface area, pore volume, CEC and more oxygen-containing groups	Wang, Gu et al. (2019)
Mg-Fe LDHs	Provide anions for co-precipitation	Adsorption of Pb ²⁺ (476.25 mg g ⁻¹)	Co-precipitation between Pb ²⁺ and interlayer anions/surface hydroxyl groups was the dominant removal mechanism	Jia et al. (2019)
Mg-Al LDHs	Promote ion exchange	Adsorption of methylene blue (406.47 mg g ⁻¹)	The adsorption process could reach equilibrium within 20 min	Meili et al. (2019)
Zn-Al LDHs	Promote ion exchange for anions	Adsorption of phosphorous (152.1 mg P g ⁻¹)	Interlayer anion exchange and complexation were fundamental adsorption mechanisms	Yang, Zhang, Sun, Tsang et al. (2019)
Mg-Al LDHs	Promote ion exchange for anions	Adsorption of phosphate (80.43 mg PO ₄ ³⁻ g ⁻¹)	Phosphate adsorption capacity increased with the increase of Mg ²⁺ /Al ³⁺ ratio owing to widened interlayer space and weakened interlayer charge density	Zhu, Hong, Ye, Hui, and Hui (2019)
Montmorillonite	Increase surface area (interlayer spaces)	Immobilization of Cu, Pb and Zn in soil	Sorption test and FTIR analysis results revealed that chemisorption was the dominant immobilization mechanism	Arabyarmohammadi et al. (2018)
Montmorillonite	Not mentioned	Adsorption of ciprofloxacin (167.36 mg g ⁻¹)	Prominent adsorption mechanisms included π - π electron donor interactions, electrophilic interactions and hydrophobic interactions.	Ashiq et al. (2019)
Montmorillonite	Provide more hydroxyl groups	Adsorption of Pb(II) (139.78 mg g ⁻¹) and atenolol (86.86 mg g ⁻¹)	Amino N and amino O generated hydrogen bonds on the surface of clay–biochar composite	Fu et al. (2020)
Montmorillonite	Increase surface area (interlayer spaces)	Adsorption of tetracycline (77.96 mg g ⁻¹)	Physisorption was the dominant adsorption mechanism	Premarathna et al. (2019)
Vermiculite	Produce Si-rich biochar	Adsorption of phosphate (49.70 mg g ⁻¹)	SiO ₂ particles on the carbon surface served as sorption sites through electrostatic interactions	Wang, Lu, Liu, and Yang (2016)
Haematite	Generate active oxygen species through Fe and S addition	Adsorption of norfloxacin (1.98 mg g ⁻¹)	Haematite addition successfully generated ·OH and SO ₄ ⁻ , promoting the degradation of norfloxacin	Yang et al. (2019)
Diatomite	Improve pore structure	Adsorption of methylene blue (153.2 mg g ⁻¹)	The composite has many pore channels in the mesoporous region (2~10 nm), favouring the adsorption of dye	Yang, Jiang et al. (2019)
Goethite	Increase surface area and promote surface complexation (formation of inner-sphere complexes)	Immobilization of Cd(II), As(III), roxarsone and phosphorous in soil	Surface complexation, ion exchange, redox reaction and co-precipitation accounted for the immobilization	Zhu et al. (2020)
Hydroxyapatite	Precipitate Pb	Immobilization of Pb in soil	The residual fraction of Pb increased by 66.6% after biochar–mineral composite addition	Yang, Fang, Tsang, Fang, and Zhao (2016)

in wastewater. The redox potential of Mn(III)/Mn(II) is higher than that of Fe(III)/Fe(II) (1.51 V and 0.77 V, respectively), indicating that in this Fenton system, Mn(III) could be reduced by Fe(II) effectively, thus breaking the limitation of H₂O₂ production on Mn(III) reduction. In this way, electron transfer in this system can be promoted, resulting in 81 times higher naphthalene decomposition rates than that of unmodified biochar (Li, Lai, et al., 2019). Graphite structures can be observed in biochars prepared under high temperatures (section 4.2.1), indicating the existence of delocalized π bonds owing to the sp² hybridization (conjugated π system) (Smith, 2016). The conjugated π electrons could migrate through Fe-O-C bonds at the interface of magnetic biochar, thus lowering the redox states of Fe species as a result of a higher electron density on the material surface. This could lead to the formation of free radicals such as $\cdot\text{OH}$ and $\text{SO}_4^{\cdot-}$ with the presence of peroxydisulphate in this system, thus promoting the degradation of sulphamethazine. Other studies have also observed that magnetic modification of graphitized biochars could promote the generation of active oxygen species with the aid of Fe(III)/Fe(II) coupling (Fu et al., 2019). A study by Yi et al. (2019) found that FeO rather than Fe₂O₃ could decompose H₂O₂ effectively through a Fenton reaction, promoting the degradation of metronidazole by OH generated through the catalytic decomposition of H₂O₂.

Several studies have also examined the performances of magnetic biochars in soil remediation. Introduction of iron oxides promoted the formation of inner-sphere complexes, resulting in reduced mobility and bioavailability of heavy metals (Wan, Li, & Parikh, 2020; Wu, Li, et al., 2020). Apart from metal stabilization, plant growth can also be promoted, but the mechanisms involved in this process remain unclear (Lu et al., 2018). It is suggested that magnetic biochars could also be used for the adsorption/degradation of organic contaminants in soil because of the generation of reactive oxygen species.

3.2 | Mineral modification

Natural minerals have been used in environmental applications for a long time owing to their environmentally benign nature and relatively low cost (Wang, Li, Tsang, Jin, & Hou, 2020). A variety of minerals can promote biochar performance, among which clay minerals (e.g., attapulgite, montmorillonite and vermiculite) have attracted much attention (Table 4) owing to their well-developed pore structures and high ion exchange capacity (Han et al., 2019). A study by Fu, Zhang, Xia, Lei, and Wang (2020) found that surface complexation between Pb(II) and hydroxyl groups provided by montmorillonite promoted the adsorption performance. It is of note that LDHs, a class of ionic lamellar compounds with

positively charged metal hydroxide layers and exchangeable anions for charge neutrality, have attracted much attention in contaminant adsorption, because LDHs possess great ability for anion exchange (Meili et al., 2019). Studies investigating the effect of alkali and alkaline earth additives on biochar production showed that the yield of carbon and, therefore, the carbon sequestration potential of biochar can be dramatically increased by addition of small amounts of these metals (Mašek et al., 2019). It is suggested that more studies of mineral modification that investigate synergies affecting carbon storage potential and environmental applications should be conducted, as this approach can be regarded as a 'green' method to improve biochar performance in a range of applications.

3.3 | Acid and alkaline activation

The aims of acid activation are to clear the pores of biochar and introduce acid binding sites (e.g., phenolic, lactonic, carboxylic functional groups) for contaminant adsorption (Li et al., 2014; Wang & Wang, 2019). A number of studies have discussed the effects of acid activation on specific surface area, pore volumes and functional groups. After activation with hydrochloric acid, sulphuric acid, citric acid, phosphoric acid or oxalic acid, the resultant biochar typically possessed much higher pore volume, surface area, more acid or hydrophobic groups for contaminant adsorption, and higher carbon retention (Hu, Xue, Long, & Zhang, 2018; Peng, Lang, & Wang, 2016; Sarkar, Ranjan, & Paul, 2019; Sun, Chen, Wan, & Yu, 2015; Wang, Ma, et al., 2019; Zhao et al., 2017). Apart from adsorption, acid activated biochar can also promote plant growth through elevating soil nutrient bioavailability. Sahin et al. (2017) modified poultry manure-derived biochar with nitric acid and phosphoric acid. Water-soluble P, K, Ca, Mg, Fe, Zn, Cu and Mn concentrations increased, thus promoting the adsorption of nutrients by maize. However, the mechanisms involved in this process were not further investigated. It is hypothesized that acid activation decreased biochar pH by introducing acid groups onto biochar surface, making the alkaline substances more labile and soluble. Acid activation could also aid in contaminant retention in soil. Vithanage et al. (2015) used sulphuric acid enhanced burcucumber biochars for sulphamethazine sorption in soil. A high solid-water partition coefficient of 229 L kg⁻¹ was observed for a loamy sand soil. Both chemisorption onto protonated functional groups and contaminant diffusion into pores were assumed to be the retention mechanisms. It is proposed that if the biomass itself contains heavy metals, acid activation may not be an appropriate modification method, as heavy metals could become more bioavailable. Heavy metals may be washed out during acid activation, and leaching tests are needed to evaluate potential changes in metal availability.

The major aims of alkaline activation are to increase specific surface area as well as the number of oxygen-containing functional groups (e.g., hydroxyl, carboxyl, carbonyl, ether) of pristine biochar, thus promoting the adsorption of a variety of pollutants (Wang & Wang, 2019). The most widely used alkaline agents are potassium hydroxide (KOH) (Bashir, Zhu, Fu, & Hu, 2018; Wang, Wang, & Wang, 2018; Wang, Ma, et al., 2019) and sodium hydroxide (NaOH) (Bogota, Sokolowska, Skic, & Tomczyk, 2019; Liu, Xue, Gao, Cheng, & Yang, 2016). After alkaline treatment, blocked pores are cleaned, resulting in greater porosity (Jin et al., 2014). Apart from contaminant adsorption from wastewater, alkaline-activated biochar can also be used as a silicon (Si) fertilizer for plant growth. Wang et al. (2018) pretreated the feedstock biomass (rice straw) using KOH (1:10 KOH: biomass wt. ratio). The KOH activation process increased plant available Si content, because higher solubility of Si under alkaline conditions increased the bioavailability of phytoliths-Si. However, KOH activation may be a double-edged sword. Wang, Bolan, Tsang, and Hou (2020) found that a low concentration of KOH (i.e., 1 M or 3 M) favoured the stabilization of Pb and Cd in soil because of clearance of blocked pores, but a high concentration (i.e., 5 M) hindered the immobilization because of the damage of the cell structure of rice husk under alkaline activation. Alkaline activation is a relatively simple modification step, since it only involves the mixing and washing processes under mild conditions. It is thus a promising and applicable modification method in environmental applications. However, the environmental footprint and economic costs need to be carefully assessed on a case by case basis because of the requirement for different chemical reagents. Additionally, relatively large volumes of water are needed for pH neutralization after acid or alkaline activation, without which the excess acidity/alkalinity may be detrimental to the environment upon biochar application.

3.4 | Oxidant modification

Oxidant modification can increase the content of oxygen-containing functional groups, promoting the complexation of heavy metals. Wang and Liu (2018) activated manure-derived biochar with hydrogen peroxide. The oxygen content and carboxyl contents of biochar increased by 63% and 101%, respectively, while the ash content decreased by 42% after activation. The activated biochar could adsorb Pb^{2+} , Cd^{2+} , Cu^{2+} and Zn^{2+} effectively, which was because of the shifting of the adsorption mechanism from precipitation to complexation. However, H_2O_2 modification was ineffective in methylene blue removal. A study by Huff and Lee (2016) found that after H_2O_2 modification of pinewood biochar, biochar methylene blue removal decreased. This was because oxygen-containing groups weakened the forces of delocalized

π interactions, which was the major mechanism of methylene blue adsorption. Apart from hydrogen peroxide, potassium permanganate could also be used as a modification agent (Wang, Dong, et al., 2019). The effectiveness of this method depends greatly on the target contaminant type and contaminant removal mechanism. It is hypothesized that this method is suitable for metal stabilization in soil (enhanced surface complexation because of an increase in oxygen-containing functional groups).

3.5 | Photocatalytic modification

Photocatalytic degradation of organic contaminants could be achieved by biochars after modification with metal oxide-based semiconductors such as TiO_2 , Cu_2O , CuO and ZnO . Doping of metal oxides on biochar can be achieved in different ways, such as the sol-gel method (Lisowski et al., 2018; Xie, Li, Zhang, Wang, & Huang, 2019), hydrolysis (Lu, Shan, Shi, Wang, & Yuan, 2019) and the hydrothermal method (Khataee et al., 2019). Lisowski et al. (2018) produced crystalline TiO_2 on biochar using a novel low-temperature ultrasound-promoted green methodology coupled with citric acid as a cross-linking agent. The resulting biochar-supported TiO_2 achieved a high phenol degradation performance even under visible light (Lisowski et al., 2017). Biochar could not only serve as a host material for metal semiconductors, but also promote the electron transfer process. For instance, Lu et al. (2019) found that the mesoporous structure of walnut shell biochar ensured the dispersion of TiO_2 nanoparticles (the role of host material). Another study by Xie et al. (2019) applied Zn- TiO_2 -loaded reed straw biochar for sulphamethoxazole degradation under visible light irradiation. Because of the electronegativity of biochar, sulphamethoxazole and intermediates come into contact with the photocatalyst more easily, and the generated electron (e^-) can then transfer to the surface of the biochar without the recombination of electron-hole pairs, thus promoting the photocatalytic process (the role of promoting electron transfer). In other words, biochar acts as an electron trapper in the conduction band of semiconductors that can accelerate electron transfer and separation of electron-hole pairs (Khataee et al., 2019).

3.6 | Electrochemical modification

Compared with other modification methods, electrochemical modification is a simple and rapid method aimed at introducing specific functional groups and impregnating chemicals onto the surface of raw biochars. For instance, Yang, Zhang et al. (2019) prepared magnetic corn straw biochar under an electric field generated by an electrode. The external electric field enabled the rod-like crystalline Fe_3O_4 nanoparticles

to disperse thoroughly into the inner pores of the biochar, resulting in a slight decrease of specific surface area (from 130 to 100 m² g⁻¹) and increase of pore diameter (from 7.56 to 7.67 nm). Electrochemical modification methods can also be used for MgO impregnation (Jung & Ahn, 2016; Jung, Hwang, Jeong, & Ahn, 2015). Using MgCl₂ as the electrolyte and graphite as the electrode, MgO nanoparticles were dispersed and enriched on the surface of biochar derived from marine macroalgae. The resulting MgO/biochar composite was shown to adsorb phosphate effectively, reaching a maximum adsorption capacity of 620 mg g⁻¹ (Jung & Ahn, 2016). MgO-doped biochars have proven to be effective for soil metal stabilization processes (Shen, Pan, et al., 2019; Shen, Zhang, et al., 2019). Electrochemical modification could be a simple method to fabricate these immobilizing agents.

3.7 | Carbonaceous nanomaterial modification

Elevated aromaticity of biochar correlates with increases in delocalized π bonds, favouring the adsorption of organic contaminants (Smith, 2016). Although a graphite-like sheet structure can be observed in biochars pyrolysed at high temperatures (>700°C) (section 4.2.1), grafting graphene oxide onto biochar is a straightforward approach to increasing the adsorption capacity of organic molecules. Graphene oxide possess a variety of oxygen-containing functional groups (e.g., hydroxyl, carbonyl, carboxyl), making the binding between graphene oxide and the host matrix (biochar) stable (Zhou, Cao, et al., 2019). The widespread sp² sheet structure indicates strong affinity with emerging contaminants with aromatic rings, such as ciprofloxacin, oestradiol and sparfloxacin (Liu et al., 2019; Zhou, Cao, et al., 2019). For instance, a study by Zhou, Cao et al. (2019) observed that the graphite structure was grown homogeneously on the surface of citrus peel-derived biochar. The graphene oxide on biochar surface served as a π electron donor, while ciprofloxacin and sparfloxacin acted as π electron acceptors owing to the inductive effect caused by the strong electron-withdrawing affinity of fluorine in the aromatic ring. Made up of multiple rolled layers of graphene, multi-walled carbon nanotube (MWCNT) is another emerging carbonaceous material with high surface area, well-developed pore structure and abundant delocalized π bonds (Duclaux, 2002). Inyang, Gao, Zimmerman, Zhang, and Chen (2014) synthesized a MWCNT/biochar nanocomposite for methylene blue removal. It has been hypothesized that electrostatic attraction (between positively charged methylene blue and deprotonated hydroxyl and carboxyl groups) as well as π - π interactions were the dominant adsorption mechanisms. It is suggested that more studies should focus on the sustainable fabrication of these nanocomposites (section 5).

3.8 | Methanol modification

A study by Jing, Wang, Liu, Wang, and Jiang (2014) modified rice husk biochar with methanol. Methanol modification could not only rinse off organic compounds blocking the pores, but increase the number of carbonyl groups on the biochar surface. X-ray photoelectron spectra (XPS) showed a shift of surface oxygen atoms from higher energy (533.5 eV) to lower energies (533.2 eV), indicating an increase in electron density of oxygen (O). The strong basicity of surface O atoms resulted in the formation of hydrogen bonds between biochar and tetracycline. In addition, the conjugated enone structures of tetracycline acted as π electron donors, thus favouring π - π interactions. However, methanol is a toxic agent, and 'greener' agents should be investigated to take the place of methanol in carbonyl introduction (section 5).

4 | EFFECTS OF FEEDSTOCK AND PYROLYSIS CONDITIONS

Both feedstock and processing conditions play important roles in the yield and properties of the final biochar products, and understanding of the relationships between these parameters is key for the production of engineered biochar.

4.1 | Feedstock

4.1.1 | Types of feedstock

Depending on the intrinsic characteristics of the feedstocks, biochar physiochemical properties may vary greatly. Selection of a proper feedstock is critical if a biochar is to be produced for some specific environmental applications.

Wood

Owing to the higher content of cellulose, hemicellulose and lignin, wood biochars have high specific surface area and low ash content (Shaheen et al., 2019). Although wood-based biochars possess similar physical and chemical properties in comparison with biochar derived from other feedstocks, biochars produced from different wood species may vary greatly. Hardwood-derived biochars had much higher alkalinity, cation exchange capacity (CEC) and micropore volume, compared with softwood-derived biochars (Jiang et al., 2017; Shaheen et al., 2019; Yargicoglu, Sadasivam, Reddy, & Spokas, 2015). In addition, the aromaticity of hardwood biochar is higher, indicating higher

stability in practical applications. This difference can likely be ascribed to the higher lignin content of hardwood compared with softwood (Shaheen et al., 2019). Therefore, care must be taken when selecting wood feedstock, to suit the target biochar application.

Crop residues

Apart from adsorption and retention of environmental contaminants, crop-derived biochar can act as a soil fertilizer as well. Li and Delvaux (2019) suggested that crop-derived biochars can be used as a Si fertilizer, since monocotyledons (e.g., rice, wheat, corn, barley and sugarcane) accumulate Si in plant tissues in the form of phytolith (McKeague & Cline, 1963; Piperno, Ranere, Holst, Iriarte, & Dickau, 2009), which is a promising Si source for plants. It is argued that in order to utilize the massive amount of crop residues most effectively, the production and applicability of crop residue-derived biochar for different purposes should be further investigated, including synergies or conflicts with alternative crop residue management strategies.

Grass

Grass can also be used as feedstock for biochar, especially certain fast-growing species. For instance, as a monocot C4 perennial grass, elephant grass grows very rapidly (four harvests a year) on different soil types. In Brazil, annual biomass production of elephant grass is 30 t ha⁻¹. Considering this feature, elephant grass has been widely used as biochar feedstock in Brazil (De Conto, Silvestre, Baldasso, & Godinho, 2016; Ferreira et al., 2019; de Jesus, Matos, Cunha, Mangrich, & Romao, 2019). In addition, producing biochar using invasive species (e.g., *Spartina alterniflora* (Saltmarsh cordgrass)) is a promising method to handle the ecological risks of biological invasion (Li et al., 2015; Luo et al., 2017).

Animal waste

Compared with plant-based biochars, biochar produced from animal waste such as poultry and swine manure possess distinct physical and chemical properties because of the very different nature of the feedstock. Notably, manure biochar has a high N content as a result of the high N content in the feedstock. If the major aim of modification is to introduce N-containing groups onto biochar surface, selecting animal waste as feedstock is therefore recommended. However, one major concern with animal waste-derived biochars is their poor stability (section 4.1.3).

Sewage sludge and anaerobic digestate

Conventional disposal methods such as incineration, sanitary landfill, application as fertilizer and anaerobic digestion have raised safety concerns, as sewage sludge itself may contain heavy metals, organic contaminants and microbial contaminants, posing risks to both humans and the environment. Pyrolysis can be a more environmentally friendly and economically acceptable approach, since it can reduce solid waste, generate fuel co-products (e.g., bio-oil) and reduce or even completely remove certain contaminants, such as microbes and antimicrobial resistance genes (Chen et al., 2014; Frišták, Laughinghouse, Packová, Graser, & Soja, 2019; Tang et al., 2018). However, care must be taken when utilizing sludge-derived biochar in environmental applications, since heavy metals are usually not completely removed during the pyrolysis process although their availability may be reduced (Lu, Yuan, Wang, Huang, & Chen, 2016; Phoungthong, Zhang, Shao, & He, 2018). Co-pyrolysis is an effective way to reduce the risks associated with the utilization of sludge-derived biochars (section 2.5). Utilization of heavy metal contaminated sludge-based biochar directly as soil amendment is not recommended, as heavy metals will mobilize with the ageing of biochar (He et al., 2019). As has been discussed in section 2.5, co-pyrolysis of sludge and other feedstocks such as crop residues and wood may reduce the risks. If the risks of metal leaching are controlled properly, or if sludge with low metal content is used (such as that from water treatment in rural areas), pyrolysis offers a promising way to handle and recycle the sludge.

Anaerobic digestion is an effective way to generate biogas from sewage sludge, crop residues, animal slurries and manure. Although the solid residue, that is digestate, can be used as soil fertilizer directly, excessive N loads from digestate have raised much concern, including the effects on plant growth, soil tilth and crop yield. In addition, environmental issues such as risks of groundwater NO₃⁻ contamination, NH₃ volatilization, N₂O emissions and pathogen exposure have restricted its applications (Monlau et al., 2016). Notably, if the aim of biochar modification is to introduce N-containing groups, it is hypothesized that using digestate as feedstock could effectively produce biochar with various N-containing functional groups on its surface through conventional slow pyrolysis methods, achieving similar results to NH₃ ambience pyrolysis with more complicated pyrolysis conditions (section 2.4). However, according to the literature reviewed, no studies have examined the feasibility of this to date.

4.1.2 | Physical properties

Biochars derived from different feedstocks possess various physicochemical properties. As is illustrated in Figure 2, the

resulting biochars may retain some feedstock features. For instance, the cell structure of original mesquite wood and corn stalk can be seen clearly in the corresponding biochar particles (Figure 2). Biochar derived from chicken manure was much smoother, and the number of pores decreased. This is consistent with the finding that specific surface area of animal waste-derived biochar was much lower than that of plant-derived biochar (Figure 5c). As for sludge-derived biochars, no obvious cell structure was observed, and the morphologies of these biochars were rougher. The specific surface area of sludge-derived biochars was the lowest (Figure 5c), because feedstock materials do not possess intrinsic pore structures.

The physical properties of biochar are dependent on the chemical composition of the feedstock. Cellulose, hemicellulose and lignin content of the feedstock will greatly influence the thermal decomposition processes, thus affecting the morphology (Jahirul, Rasul, Chowdhury, & Ashwath, 2012). Thermal stability of these components follows the order of hemicellulose < cellulose < lignin (Jahirul et al., 2012). Thus, a higher lignin content (e.g., in wood) results in more ordered structure of biochar, as compared with crop or grass-derived biochar.

4.1.3 | Chemical properties

Proximate analysis of biochar reveals ash, volatile matter and fixed carbon content. As shown in Figure 3, proximate properties of biochars prepared from different feedstocks vary greatly. For instance, sludge-derived biochar has higher ash content (>50%), while ash content in wood-derived biochar is the lowest (<20%). Fixed carbon content in animal waste-derived biochars was similar (within the range of 20%–35%), while ash and volatile contents in these biochars were quite different. Ash content in biochar is highly dependent on the compositional chemistry of the initial feedstock and follows the order of sludge > animal waste > crop residues > wood. This is because inorganic mineral components are mostly retained during pyrolysis, and higher feedstock ash content results in higher biochar ash content (Singh, Singh, & Cowie, 2010).

Stability is a critical feature of biochar in environmental applications and is highly dependent on feedstock. It has been acknowledged that higher lignin content results in higher aromatic carbon (C) content, leading to higher stability (Leng & Huang, 2018). Because of this, wood-derived biochars possess higher stability than sludge, grass and crop-derived biochars. Higher

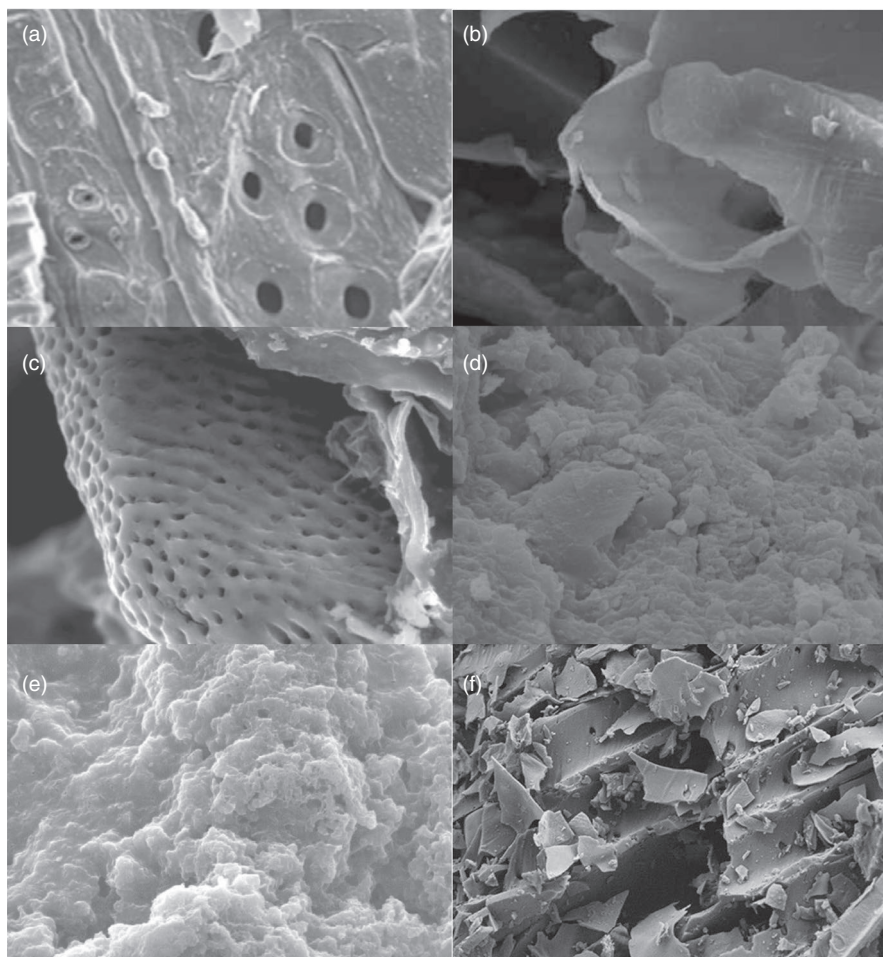


FIGURE 2 Morphology of biochar produced from different feedstocks. (a) mesquite wood (Trigo, Cox, & Spokas, 2016); (b) corn stalk (Ma, Zhao, & Diao, 2016); (c) chicken manure (Joseph et al., 2010); (d) sewage sludge (Song, Xue, Chen, He, & Dai, 2014); (e) anaerobic digestate (Tang et al., 2019); (f) coconut husk (Suman & Gautam, 2017)

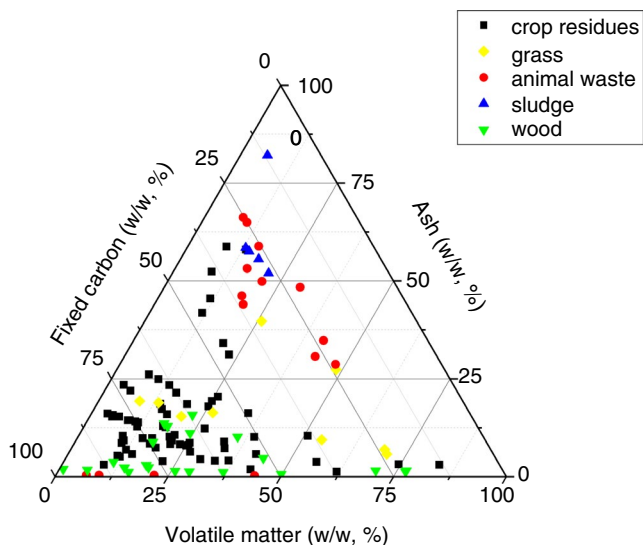


FIGURE 3 Triangle plot of volatile matter, ash and fixed carbon contents in various types of biochars. Data retrieved from: Ahmad et al., 2012; Ameloot, Graber, Verheijen, & De Neve, 2013; Angin & Sensoz, 2014; Aslam, Khalid, Naveed, & Shahid, 2017; Bian et al., 2019; Burhenne, Damiani, & Aicher, 2013; Cantrell, Hunt, Uchimiya, Novak, & Ro, 2012; Chen & Chen, 2009; Chen, Zhou, & Zhu, 2008; Chen et al., 2014; Heitkötter & Marschner, 2015; Jia et al., 2018; Jiang et al., 2015; Jimenez-Cordero, Heras, Alonso-Morales, Gilarranz, & Rodriguez, 2013; Johari et al., 2016; Karaosmanoğlu, Işigigür-Ergüdenler, & Sever, 2000; Keiluweit et al., 2010; Khanmohammadi, Afyuni, & Mosaddeghi, 2015; Klasson et al., 2014; Kloss et al., 2012; Lee et al., 2010, 2013; Li, Wang et al., 2018; Lian, Huang, Chen, Xing, & Zhu, 2011; Mohan et al., 2007; Oh & Seo, 2015; Rajarao, Mansuri, Dhunna, Khanna, & Sahajwalla, 2014; Ro, Cantrell, & Hunt, 2010; Song et al., 2014; Speratti, Johnson, Sousa, Dalmagro, & Couto, 2018; Štefelová, Zelenka, & Slovák, 2017; Suliman, Harsh, Fortuna, Garcia-Perez, & Abu-Lail, 2017; Tang et al., 2019; Tong, Li, Yuan, & Xu, 2011; Tran, You, & Chao, 2016; Trigo et al., 2016; Ucar & Ozkan, 2008; Uchimiya, Wartelle, Klasson, Fortier, & Lima, 2011; Vithanage et al., 2018; Xian et al., 2018; Xiao, Xue, Gao, & Mosa, 2017; Yuan et al., 2013; Zhao et al., 2018; Zhou et al., 2018

aromaticity indicates lower bioavailability for microorganisms to degrade biochar. The aromatic C in biochar mainly comes from the alteration from O-alkyl C to aryl and O-aryl furan-like structures, which can be concluded from the diffuse reflectance infrared Fourier transform spectroscopy (DRIFT) and solid-state ^{13}C nuclear magnetic resonance (NMR) analyses (Baldock & Smernik, 2002).

4.2 | Pyrolysis conditions

4.2.1 | Pyrolysis temperature

Pyrolysis temperature has been acknowledged as the most pivotal factor affecting biochar characteristics, and a number

of studies have investigated its influence on biochar yield, and physical and chemical properties. The molecular structures of biochar produced under different temperatures are presented in Figure 4. When the pyrolysis temperature is relatively low, the intrinsic structure of biomass materials (i.e., amorphous lignin, amorphous hemicellulose and crystalline cellulose) was preserved, and the dominant reaction at this stage is dehydration (Liu & Han, 2015). With the increase of pyrolysis temperature, biomass undergoes both depolymerization and dehydration processes, while depolymerization products of cellulose and lignin (i.e., aldehyde, carboxyl and ketones) can be observed (Keiluweit, Nico, Johnson, & Kleber, 2010). If pyrolysis temperature increases, the resultant biochar tends to reveal amorphous characteristics. At this stage, cellulose is completely depolymerized, and the proportion of aromatic lignin residues increases (Collard & Blin, 2014; Keiluweit et al., 2010). At relatively high temperatures, turbostratic crystallites are observed, and graphene-like sheets grow (Berhanu et al., 2018).

Pyrolysis temperature affects biochar yield, pH, CEC, pore structures and other physiochemical properties greatly. As shown in Figure 5a, biochar yield decreases with increasing pyrolysis temperature, and this trend is most obvious for wood-derived biochars and in the temperature range 300 to 500°C. As shown in Figure 5b, a slight increase in biochar pH is observed. This is because weak bonds in biochars (e.g., hydroxyl bond) are broken within the biochar structure at higher temperatures (Li, Barreto, Li, Chen, & Hsieh, 2018). Although most biochars are alkaline, biochar with pH < 7 can be produced at lower pyrolysis temperatures (e.g., <400°C). Therefore, if biochar is to be applied for soil pH mediation, selection of a proper pyrolysis temperature is crucial. Pyrolysis temperature also affects morphological properties (Figure 5c). With the increase of pyrolysis temperature, the specific surface area of biochars increases greatly. It is hypothesized that decomposition of aliphatic carboxyl and alkyl groups and the exposure of lignin aromatic cores are responsible for this increase (Ahmad et al., 2014; Chen & Chen, 2009). However, the surface area and pore space of the resultant biochar tend to decrease at higher pyrolysis temperature (i.e., >700°C) (Figure 5c). This is because of unblocking of micropores (Ahmad et al., 2014; Keiluweit et al., 2010). Different pyrolysis temperatures will result in various proximate properties. In general, a reduction of volatile matter can be observed with increasing temperature (Figure 5d). This is because at higher temperatures more labile forms of C (i.e., volatile matter) are released (Xie et al., 2015).

Ultimate properties and biochar stability change greatly with the increase of pyrolysis temperature (Figure 6). A reduction of hydrogen (H) and O content, and subsequently of H/C and O/C atomic ratios, indicates dehydration and deoxygenation of the feedstock materials, together with higher aromaticity and lower polarity of the resultant biochars (Ullah et al., 2019). This is ascribed to an increasing degree

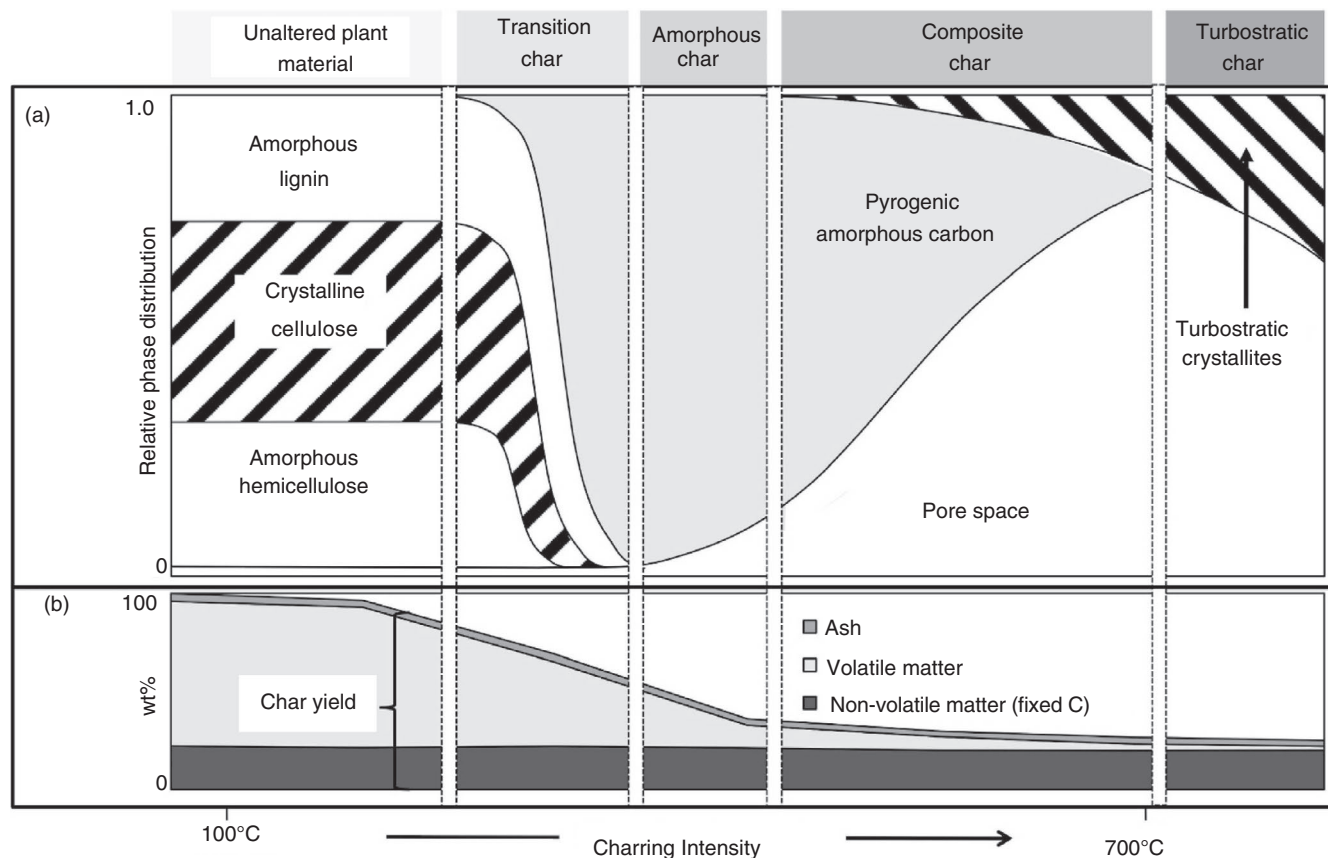


FIGURE 4 Dynamic molecular structure of plant biomass-derived biochar across a charring gradient. Reprinted with permission from Keiluweit et al., 2010. Copyright 2010 American Chemical Society

of carbonization with an elevation in charring temperature (Kim, Kim, Cho, & Choi, 2012). Graphite is regarded as a stable C form with low O content. Spokas (2010) suggested that a low O/C value (i.e., <0.2) will lead to long-term stability (half-life > 1,000 years). It is of note that the aim of several novel pyrolysis and modification strategies is to increase biochar aromaticity, promoting biochar adsorption of organic contaminants through π - π interactions (sections 2.1 and 3.7). A higher pyrolysis temperature aligns with this goal (Figure 1).

4.2.2 | Heating rate

Depending on the heating rate, conventional pyrolysis methods can be divided into two broad categories, namely slow pyrolysis and fast pyrolysis. Slow pyrolysis, the most widely used pyrolysis method, has a long history of being utilized for charcoal production. A lower heating rate provides a milder condition for the pyrolysis reactions to complete, and suitable conditions for secondary char formation to take place, which favours the formation of carbonaceous biochar (Huang, Kudo, Masek, Norinaga, & Hayashi, 2013; Keiluweit et al., 2010). During fast pyrolysis, biomass is

heated at a much higher heating rate ($>10^{\circ}\text{C s}^{-1}$). Fast pyrolysis favours the production of bio-oil and pyrolysis gas rather than biochar. Before the biomass can be decomposed into solid biochar, it is instead converted into a liquid product. The biochar yield is usually below 20%, while bio-oil yield is typically above 50%.

Even within the individual categories, for example slow pyrolysis, heating rates can vary in magnitude and this affects the yield and resulting biochar properties. It is thought that higher heating rates lead to partial graphitization, thus decreasing the surface area (Fu et al., 2012). Heating rate also influences the surface morphology of biochars. Cetin, Gupta, and Moghtaderi (2005) observed that a high heating rate (up to $500^{\circ}\text{C s}^{-1}$) led to the loss of cell structure and natural porosity of radiata pine biochar, which was because of plastic transformations (i.e., melting of cell structures).

With the exception of small batch reactors with appropriate instrumentation, heating rate is often not measured directly, but is inferred based on other measurements. This becomes especially complex in continuous pyrolysis reactors with complex geometries and material flows. Therefore, reporting of details of such measurements and calculations used becomes critical to enabling reliable comparisons of data from different studies.

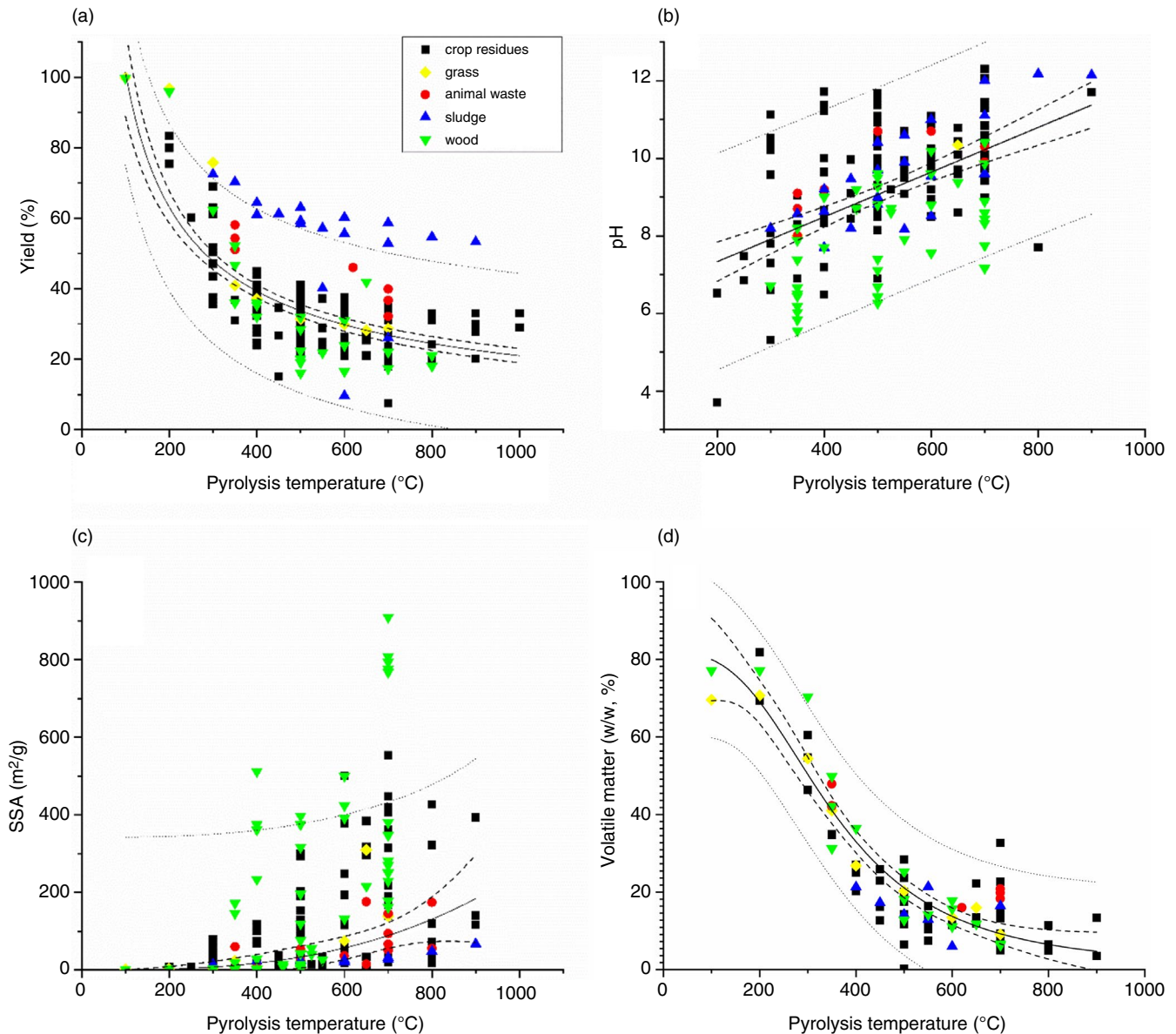


FIGURE 5 Relationships between biochar yield, pH, specific surface area (SSA), volatile matter and temperature

4.2.3 | Residence time

Residence time is another criterion separating slow and fast pyrolysis approaches (minutes to hours vs. several seconds). It affects both carbonization rate and yield of biochar. A longer residence time usually results in enhanced biochar carbonization, leading to lower labile carbon content (which has a lower microbial bioavailability) (Zornoza, Moreno-Barriga, Acosta, Munoz, & Faz, 2016). In addition, if biochars were produced at a relatively low temperature (<500°C), extending the residence time leads to an elevated ash content and decreased H and N content owing to burning-off of organic matter (Mui, Cheung, Valix, & McKay, 2010). At higher pyrolysis temperatures, the higher decomposition rate of polymers (i.e., cellulose, hemicellulose and lignin) indicates that

holding time is a less critical parameter, since the depolymerization and carbonization reactions can go to completion in a relatively short time (Cross & Sohi, 2013; Li, Amin, et al., 2019).

Residence time also affects the physical properties of biochar. Longer holding time results in the enhanced formation of both micro- and macropores, thus increasing the specific surface area. When considering the effects of residence time, it is important to distinguish between the different meanings/terminology often used. Residence time can refer to the overall residence time of a material in a reactor, or to the residence time at the peak temperature. In continuous systems, different particles of biomass/biochar will experience different residence times, and therefore, residence time distribution is an important parameter to consider (Masek et al., 2018). Often

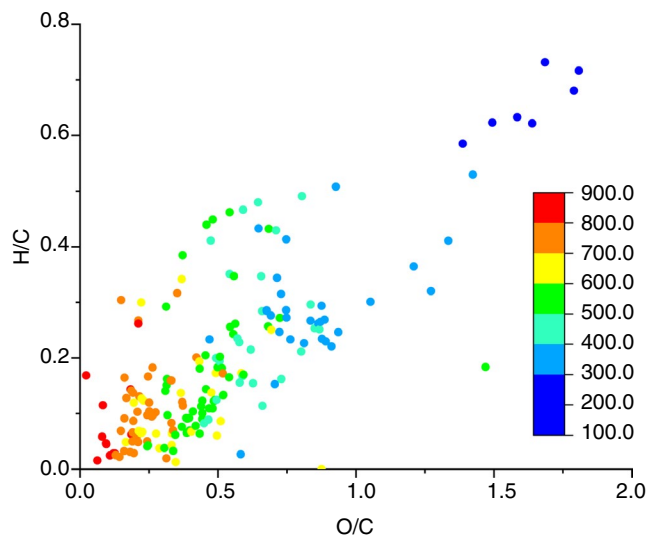


FIGURE 6 Van Krevelen diagram showing the effect of pyrolysis temperature on aromaticity (colder colours reflect lower pyrolysis temperature, while warmer colours represent higher pyrolysis temperature)

insufficient information is provided in publications to make it clear which residence time is being referred to and how it was determined. It is also important to recognize that the residence time for biochar, vapours and gases will be different. As a result, it is often difficult to draw a straightforward conclusion about the role of residence time on biochar properties.

Since biochar physiochemical properties are greatly affected by feedstock and pyrolysis conditions, it is suggested that in environmental applications, selection of a proper feedstock and pyrolysis parameter can assist in biochar modifications (Figure 1). For instance, if the aim of biochar modification is to achieve high specific surface area and pore volume, selecting a longer residence time and relatively high pyrolysis temperature will aid in steam-assisted pyrolysis or acid/alkaline modification. If the aim is to introduce more O-containing active sites for surface complexation, choosing a relatively low pyrolysis temperature and lower heating rate is favourable prior to oxidant or methanol modifications. If the target of modification is to introduce more N-containing functional groups (i.e., NH_3 ambiance pyrolysis), using N-rich feedstocks such as anaerobic digestate and animal waste may be helpful. It is suggested that future studies should systematically assess the effects of feedstock and pyrolysis parameters on biochar characteristics before the application of modification techniques.

5 | SUSTAINABILITY CONSIDERATIONS

Various stakeholders including researchers, practitioners, regulators and the public are calling for increased

sustainability to minimize life cycle environmental footprints and maximize social and economic benefits (Hou, 2020; Hou & Al-Tabbaa, 2014; Jia et al., 2020; Wang, Hou, et al., 2019). To ensure the cost-efficiency of biochar, selection of pyrolysis strategies should first focus on the major aims of biomass treatment. Although a variety of novel pyrolysis methods have been proposed, a cost-competitive biochar pyrolysis process is what the market really needs to adopt these products. Compared with fast pyrolysis and flash pyrolysis, slow pyrolysis is recommended if biochar production is the major aim, owing to higher biochar yield and biochar quality (Hersh, Mirkouei, Sessions, Rezaie, & You, 2019). Modification of slow pyrolysis processes with the aid of microwave, steam or NH_3 could improve biochar properties greatly, resulting in enhanced performances in environmental applications. However, if the major aims of biomass pyrolysis are energy applications (i.e., pyrolysis gas or bio-oil), fast pyrolysis can be adopted, with biochar as a by-product of this process (Dai et al., 2017).

When it comes to biochar modification, green methods without the use of toxic agents are highly recommended. Mineral modification, acid/alkaline modification and magnetic modification are encouraged, since the agents used in these processes are either natural or non-toxic. However, some novel modification strategies involve the utilization of toxic agents, which may raise occupational safety risks. For instance, although methanol can introduce carbonyl groups successfully onto the biochar surface, this chemical can destroy the function of optic nerve (10 ml pure methanol ingestion could cause permanent blindness) (Vale, 2007). As a second example, recent studies have examined the feasibility of using biochar-supported nanoscale zero-valent iron (nZVI) for catalytic degradation of trichloroethylene (Li et al., 2020) and adsorption of metals (Yang et al., 2018). Toxic sodium borohydride (NaBH_4) was used in the reduction of Fe(II) to nZVI, and the associated risks were overlooked. Although grafting graphene or C nanotubes onto biochar surface could effectively promote biochar's affinity towards organic contaminants with aromatic rings, the complicated procedure of nanocomposite generation may hinder its practical adoption to niche applications. It is argued that more green synthesis and modification methods of engineered biochar production be proposed to minimize the environmental impact and costs, resulting in higher 'net environmental benefit' (Wang, O'Connor, et al., 2019).

Notably, if feedstock itself contains contaminants (e.g., certain types of sewage sludge, demolition wood, phytoremediation biomass), the associated risks of contaminant release should not be neglected. Co-pyrolysis of contaminated feedstock and uncontaminated biomass is a feasible method to reduce the potential risks of contaminant leaching. There is a trend that plastics have been applied in co-pyrolysis processes to increase hydrophobicity and promote electrostatic

interactions between biochar sorbent and contaminants in wastewater. However, recent studies have also found that under certain circumstances co-pyrolysis of biomass with plastics can increase the risk of release and migration of microplastics because of plastic materials breaking into smaller pieces during pyrolysis (O'Connor et al., 2019; Oh & Seo, 2019). Reducing the risks from traditional contaminants may result in elevated risks from emerging contaminants such as microplastics. This should be avoided by taking care when selecting feedstock materials and pyrolysis conditions. Research in this area is very limited to date, and a concentrated research effort is encouraged to establish safe practices and operational boundaries.

6 | FUTURE RESEARCH DIRECTIONS

It is without doubt that biochar has emerged as an important potential tool in environmental applications. To facilitate further applications of biochar, there are several issues that should be investigated.

The results from wastewater treatment should be extrapolated to soil remediation/fertility improvement. The similar aims of the modification and remediation mechanisms will be the key to this expansion.

More in situ or field experiments should be conducted on a longer term to simulate the actual environment and examine the real effect of biochar prior to large-scale applications.

More studies should investigate the associated risks of biochar derived from different types of sludge or other contaminated biomass, and effects of various means to reliably reduce the risks (e.g., effects of co-pyrolysis).

More systematic studies should be conducted to investigate the relationship between feedstock/pyrolysis conditions and biochar physiochemical properties in more depth (e.g., the relationship between biochar structure and stability; the role of cellulose, hemicellulose and lignin content), and novel statistical analysis methods such as machine learning can be adopted to 'predict' biochar properties. For instance, Zhu, Li, and Wang (2019) used a random forest data mining method to predict the biochar yield and carbon contents. It is suggested that statistical analysis results should be further mined for information, and the underlying mechanisms/relationships should be discussed.

More attempts should be made to further expand the modification methods and environmental applications of engineered biochar with the aim of simplifying the modification steps, reducing the cost and maximizing the applicability of a certain type of engineered biochar to assure sustainability.

Future studies should focus on the mechanisms involved in environmental applications of modified biochars. For instance, although several studies have proven that acid or alkaline-activated biochar could promote plant growth, the

mechanisms involved and govern those processes are not well understood.

More attempts should be made to examine the feasibility of combining novel pyrolysis methods with emerging modification strategies to produce more effective biochars (e.g., co-pyrolysis and magnetic modification to produce sludge-derived magnetic biochars with low environmental risk; microwave-assisted pyrolysis and alkaline activation to produce biochars with extremely high specific surface areas approaching that of activated carbon).

7 | CONCLUSIONS

Various biochar modification methods aimed at improving the pore structure, increasing certain functional groups, promoting the generation of activated oxygen species and reducing risks associated with contaminants have been proposed and tested. Two main categories have emerged, namely modification of biochar postpyrolysis and modification of pyrolysis conditions. Recent literature suggests there are several new trends in biochar pyrolysis and modification strategies:

Firstly, green synthesis and modification methods are emerging to enhance sustainability, and non-toxic materials such as minerals and metal oxides have emerged as novel modification agents. Secondly, the combination of biochar with other novel materials (e.g., nanofibers, graphene, LDHs, MWCNTs) has emerged as a new approach to improving biochar characteristics, and various strategies such as microwave-assisted pyrolysis and co-pyrolysis can graft these materials onto biochar successfully. Thirdly, reducing the risks of using contaminated biomass-derived biochars in environmental applications could be achieved through new methods such as co-pyrolysis. Finally, the application of certain modification methods has been expanded. For instance, although magnetic modification was originally used for promoting biochar adsorption, recent studies have found that it could also be utilized for the catalytic degradation of organic contaminants. KOH-activated biochar can not only be used for adsorption, but also as a soil fertilizer owing to its high Si content (if the feedstock is Si-rich). Co-pyrolysis can not only reduce environmental risks of biochars pyrolysed from metal-rich feedstocks, but may also act as a new method to synthesis nanocomposites of biochar and other emerging materials such as layered double hydroxides, as well as increasing biochar's carbon sequestration potential. As a soil amendment with a long history, this carbon-rich material has revealed new vitality with the help of novel pyrolysis and modification strategies.

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