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REVIEWS AND ANALYSES

Biochar application in biofiltration systems to remove nutrients, pathogens, and pharmaceutical and personal care products from wastewater

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Abstract

Although conventional on-site wastewater treatment systems (OWTSs) provide only primary treatment of domestic wastewater, removal of a limited level of nutrients (N, P), pathogens, and pharmaceuticals and personal care products (PPCPs) could be achieved by such a treatment process. Biochar has the capacity to remove various contaminants and has been widely used as an ideal soil amendment in agriculture due to its persistence, superior nutrient-retention properties, low cost, and ready availability. However, few applications on the use of biochar in onsite wastewater treatment have been explored. In this review, we systematically reviewed the applications of biochar in filtration-based OWTSs for nutrient (N, P) removal and recovery as well as pathogen and PPCP removal. Although adsorption was the main mechanism for P, pathogen, and PPCP removal, biochar can also serve as the growth media for enhanced biological degradation, improves available alkalinity, and increases water holding capacity in the OWTSs. The biochar source, surface modification methods, and preparation procedures (e.g., pyrolysis temperature change) have significant effects on contaminant removal performance in biochar-amended OWTSs. Specifically, contradictory results have been reported on the effect of pyrolysis temperature change on biochar removal performance (i.e., increased, decreased, or no change) of N, P, and PPCPs. Wastewater composition and environmental pH also play important roles in the removal of nutrients, pathogens, and PPCPs. Overall, biochar holds great potential to serve as an alternative filtration material or to be amended to the current OWTS to improve system performance in removing a variety of contaminants at low cost.

Abbreviations: CEC, cation exchange capacity; CW, constructed wetland; HRT, hydraulic retention time; NP, nanoparticle; OWTS, on-site wastewater treatment system; pH_{pzc}, pH at the point of zero charge; PPCP, pharmaceuticals and personal care product; STE, septic tank effluent; SSA, specific surface area; SWIS, subsurface wastewater infiltration system; TN, total nitrogen; TP, total phosphorous; WHC, water holding capacity; ZVI, zerovalent iron.

1 | INTRODUCTION

More than 25 million households in the United States and hundreds of millions of households around the world rely on on-site wastewater treatment systems (OWTSs) to treat sewage before discharging it to the environment (Amador & Loomis, 2019; Petitjean et al., 2016). Domestic on-site wastewater contains a variety of contaminants, including organic matter (i.e., biochemical oxygen demand), nutrients (nitrogen [N] and phosphorous [P]), pharmaceutical and personal care products (PPCPs), and pathogens (i.e., bacteria and viruses) (Farkas et al., 2020; Guruge et al., 2019; Martikainen et al., 2018). The conventional OWTS, which is comprised of a septic tank followed by a drain field or a leaching pool (USEPA, 2002b), can provide a limited level of nutrient and pathogen removal, depending on the soil characteristics (Amador & Loomis, 2019). On-site wastewater is one of the largest sources of legacy N in soil, and the excess nutrients released into shallow groundwater threaten drinking water quality and are directly linked to eutrophication and harmful algal blooms (Kinney & Valiela, 2011; Meter et al., 2018; Shahraki et al., 2020; Wolfe & Patz, 2002). The composition of the wastewater stream (i.e., septic tank effluent [STE]) fluctuates significantly in an OWTS because many factors (e.g., weather conditions, the number of people living on site, and the water use pattern) affect contaminant levels. Compared with the municipal wastewater treatment plants, the levels of total N (TN) (42.8–93.5 mg N L^{-1}), alkalinity (150–330 mg $CaCO_3$ L^{-1}), total P (TP) (3.0–9.5 mg L^{-1}), and total coliform (1.5×10^6–1.9×10^7 most probable number 100 ml^{-1}) varied significantly in the STE at different sites (Gobler et al., 2021; Richards et al., 2017).

Compared with the conventional activated sludge–based OWTSs, soil-based biofiltration systems are selected as advanced OWTSs in many areas due to their nutrient removal capacity and their low construction and maintenance costs (Cooper et al., 2015). Constructed wetlands (CWs), N-removing biofilters, and subsurface wastewater infiltration systems are common soil-based advanced OWTSs. Constructed wetlands have been developed based on the natural wetlands with mechanical units (e.g., aeration unit and flow distribution unit) and have been used for wastewater treatment processes (H. Wu et al., 2015). Nitrogen-removing biofilters are engineered biofiltration systems that consist of (a) an unsaturated top sand layer (aerobic) for biological oxygen demand removal and nitrification and (b) a bottom saturated lignocellulose/sand layer (anaerobic) for heterotrophic denitrification. These systems have been reported to be effective in nutrient (N and P) removal from onsite wastewater (Waugh et al., 2020; Wehrmann et al., 2020). The subsurface wastewater infiltration system (SWIS) contains a wastewater distribution unit and a selected type of media, such as soil and gravel, below the distribution unit (C. Liu et al., 2018). A biomat is formed at the wastewater–soil interface, and the contaminants are degraded in the unsaturated soil matrix (USEPA, 2002b; X. Wang et al., 2010).

Core Ideas

- Biochar in on-site wastewater treatment systems removes pollutants via multiple mechanisms.
- Pyrolysis condition affects biochar properties and its contaminant removal capacity.
- Biochar can recover phosphorus from on-site wastewater and be used as a soil fertilizer.
- Biochar in biofiltration systems can enhance pathogen removal from on-site wastewater.
- Modified biochar increases the removal of pharmaceutical and personal care products.

Various types of media have been amended to soil-based biofiltration systems for wastewater treatment (e.g., oyster shell, limestone, and zeolite) to improve contaminant removal (Gill et al., 2009; Gungor & Unlu, 2005; Z. Wang et al., 2013; Y. Zhang et al., 2015). Among different amendment materials, biochar holds great potential to enhance contaminant removal due to various mechanisms (Mohanty et al., 2018). Biochar is the solid material obtained from thermochemical conversion of biomass in an oxygen-limited environment (IBI, 2015). Biochar can adsorb various organic compounds, increase the cation exchange capacity (CEC), and increase the matrix pH level; therefore, the metal precipitation process is enhanced with the abundant organic functional groups on the biochar surface (Fidel et al., 2017; Oliveira et al., 2017). The biochar structure is highly porous and can therefore provide high surface area (up to 635 m^2 g^{-1}) and high water holding capacity (up to 2.5–2.9 ml g^{-1}) (Lehmann & Joseph, 2015; Rajkovich et al., 2011). In addition, biochar has the potential to manipulate redox conditions by acting as an electron acceptor or electron donor during microbial degradation processes (Saquing et al., 2016). Furthermore, biochar can serve as the growth media for biofilm, enhancing the microbial abundance and biological activity (Bock et al., 2015, 2016; He et al., 2018b; Liang et al., 2020). On the other hand, the biochar source and production conditions (e.g., temperature, residence time of pyrolysis, gas flow rate, and additives) can significantly change the physical and chemical properties of biochar (Oliveira et al., 2017). Due to its unique physical and chemical characteristics, low cost, and ready availability, biochar has been widely used as an ideal soil amendment in agriculture to enhance crop yield by acting as a soil amelioration material and as a slow-releasing nutrient source (Lehmann & Joseph, 2015). Biochar is also efficient in removing heavy metals and organic compounds and in electrochemical treatment

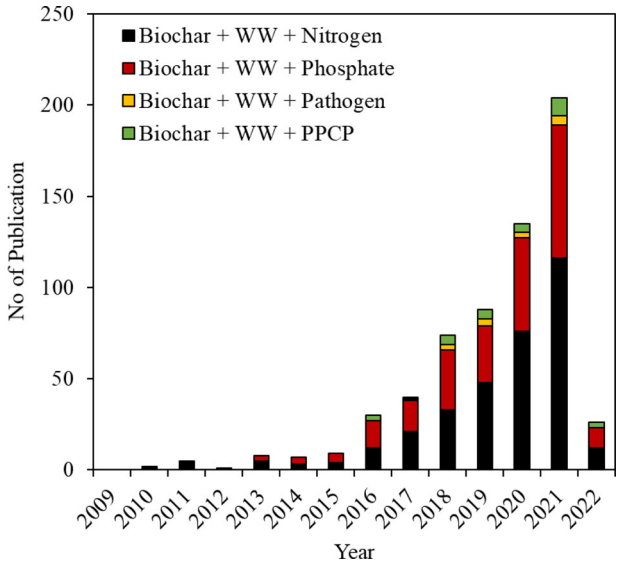

FIGURE 1 Number of publications related to biochar and wastewater based on Web of Science (last updated in January 2022). PPCP, pathogens, pharmaceuticals, and personal care product; WW, wastewater

applications (Q. Huang et al., 2019; Inyang et al., 2015; Z. Wu et al., 2019).

Interest in using biochar for removal of nutrients, pathogens, and PPCPs during the wastewater treatment process has increased significantly in the past decades. The combination of biochar with biofiltration for domestic wastewater treatment has been suggested (Xiang et al., 2020). In on-site wastewater treatment areas, biochar may serve as a sustainable and effective amendment to the soil-based OWTSs to enhance system performance in removing a variety of contaminants (Enaime et al., 2020). In this study, we conducted a comprehensive review on (a) the main mechanisms involved in removal and recovery of nutrients, pathogens, and PPCPs when biochar was amended in the biofiltration-based OWTS and (b) factors that affect biochar-amended biofiltration-based OWTS performance on the removal of nutrients (N and P), pathogens, and PPCPs. The challenges and future studies that are needed for the wide application of biochar in on-site wastewater treatment are also discussed.

2 | REVIEW SEARCH CRITERIA

For this review, a systematic literature search of the peer-reviewed publications in Web of Science was conducted to investigate the biochar application in biofiltration systems with the purpose of evaluating their application in onsite wastewater treatment. The search terms included "wastewater," "biochar," "nitrogen," "phosphate," "pathogen," and "PPCPs." The literature search was limited to peer-reviewed publications written in English and published mostly between 2009 and 2022 (last update on 18 Jan. 2022) (Figure 1). Approximately 630 results can be searched from the database using different combinations of the above keywords. After a full-text review, 190 references, including primary research articles, reviews, and book chapters, passed our criteria. Study inclusion criteria, including scope and data availability, were applied to each publication. Studies on the use of biochar in the removal of metals and organic pollutants from domestic and industrial wastewater are excluded because they are beyond the scope of this review. This review provides up-to-date information regarding the applications of biochar in biofiltration systems with the intent of use as onsite wastewater treatment, the removal/recovery mechanisms, critical factors that affect the system performance, and future directions of biochar research in onsite wastewater treatment areas.

3 | APPLICATION OF BIOCHAR IN BIOFILTRATION SYSTEMS FOR N REMOVAL

3.1 | N removal mechanisms

3.1.1 | Adsorption

The microporous structure and the various organic functional groups (e.g., carbonyl, carboxyl, phenolic hydroxyl, and hydroxyl groups) on the biochar surface provided the adsorption and ion exchange potential for nutrient removal (Lehmann & Joseph, 2009; Yue et al., 2017). For example, acidic functional groups (e.g., hydroxyl, phenol, and carboxyl groups) and basic organic functional groups (e.g., amides, aromatic amines, and pyridinic groups) provide cation and anion exchange sites for NH_4^+ and NO_3^- present in the wastewater stream (Q. Yin et al., 2017). In unmodified biochar, the adsorption capacity of NH_4^+ (0.7–17.6 mg NH_4^+ N g^{-1}) was reported to be much higher than NO_3^- (0.7–2.8 mg NO_3^- N g^{-1}) when the influent contained 10–100 mg NH_4^+ N L^{-1} and 10–20 mg NO_3^- N L^{-1} (Q. Yin et al., 2017). The difference in adsorption capacity was mainly due to the abundant acidic functional groups on the biochar surface. Both Freundlich (i.e., multilayer) and Langmuir (i.e., monolayer) adsorption have been reported in NH_4^+ adsorption tests with both modified and unmodified biochar (Vu et al., 2017; H. I. Yang et al., 2018; Q. Yin et al., 2018). At neutral pH (~7), biochar showed effective removal of NH_4^+, with a pH at the point of zero charge (pH_{pzc}) of 5.6–5.8, indicating that the negative surface charge of biochar was beneficial for NH_4^+ removal (C. Wang et al., 2020; Xiao et al., 2020). No NO_3^- adsorption (1.5 g biochar in 30 ml) was observed in batch adsorption tests, and NO_3^- was even released to the solution (0.16–0.40 mg N g^{-1}) (Gai et al., 2014). In a case study, the

surface area of biochar was found to be the key factor that controls NO_3^- adsorption, and the Langmuir model fitted better to the adsorption isotherms, indicating that NO_3^- adsorption could be a physical adsorption process in the tested biochar (J. Yang et al., 2017).

Cation exchange capacity was proven to be an important factor controlling N removal in biochar-amended biofilters (Rahman et al., 2021). Because ammonium acetate was used in the standard CEC measurement (USEPA, 1986), it is challenging to distinguish the contribution of adsorption and/or cation exchange to NH_4^+ removal by the biochar. A higher NH_4^+ removal rate (2.7 mg N g^{-1}) was observed in a biochar-amended stormwater bioretention system compared with a sand system (0.04 mg N g^{-1}), which was largely due to the higher CEC of the biochar (10.6–13.6 $cmol_c$ kg^{-1}) (Yeasir et al., 2020).

3.1.2 | Alkalinity supplement

The alkalinity provided by biochar can be categorized in four major types: (a) surface organic functional groups, such as hydroxyl and carboxyl groups; (b) soluble organic compounds (i.e., conjugate bases of acidic groups) released by biochar; (c) carbonates (i.e., salts of bicarbonate and carbonate); and (d) other inorganic alkalinities, such as oxides, hydroxides, sulfates, sulfides, and orthophosphates (Fidel et al., 2017). The soluble organic and inorganic alkalis can provide short-term buffering capacity, whereas the surface organic functional groups may provide long-term buffering capacity (Fidel et al., 2017). For example, biochar generated from chicken litter provided a buffering capacity of 62.5 mmol H^+ kg^{-1}, which is three times higher than the buffering capacity provided by the soil matrix (17.9 mmol H^+ kg^{-1}) (Palanivell et al., 2019). In the same study, with 20% (w/w) biochar amendment, the soil buffering capacity increased to 31.3 mmol H^+ kg^{-1} (Palanivell et al., 2019).

Biological nitrification is an alkalinity-consuming process, and the pH in OWTSs may decrease to as low as 3.7–4.0 (Maleki Shahraki et al., 2021). Low pH in the nitrification layer could impair the overall N removal process because the optimum pH for nitrification is 7.0–8.0 (USEPA, 2002a). Therefore, biochar amendment in the biofiltration system may facilitate sustainable nitrification by providing additional alkalinity. For instance, when biochar was amended to the phenol-contaminated wastewater, it neutralized acidic intermediates generated during the treatment process and subsequently reduced the pH shock to the microbial community (Zhao et al., 2020). In agricultural applications, biochar generated from agricultural waste (e.g., corn stover, canola stover, peanut stover, and rice straw) was reported to enhance the soil pH buffering capacity by 85–200% when 5% (w/w) biochar was amended to the soil matrix (Shi et al., 2019, 2017). In a long-term experiment, Dai et al. (2014) found when 3% (w/w) biochar was applied to the soil matrix, pH increased one to two units (from 4.0–5.0 to 5.5–7.5) during the 180 d of incubation.

3.1.3 | Water holding capacity increase

Water holding capacity (WHC) is the amount of water that can be stored in the filtration matrix (Piedallu et al., 2011). Biochar can increase the WHC of filtration media due to its large surface area and very porous structure (Lehmann & Joseph, 2009). Increased WHC may subsequently increase the hydraulic retention time (HRT) in the filtration system. For example, an 18% (v/v) biochar amendment (feedstock: oak and cedar wood; pyrolysis temperature: 550 ˚C) to the pilot-scale bioretention system increased the system WHC by 11–27%, and the HRT increased from 4.4–7.1 to 7.1–8.0 h with the same hydraulic loading (5.5 cm h^{-1}) (Tian et al., 2019). In another study, 5% (w/w) biochar amendment (feedstock: sawdust; pyrolysis temperature: 700 ˚C) to the soil matrix increased the WHC up to 2–3 g water g^{-1} soil, which was almost three times higher than the soil matrix without biochar amendment (Mao et al., 2019). When treating domestic wastewater, increased HRT in CWs (from 4 to 8 d) was reported to result in better NH_4^+ (from 44.4 to 59.3%) and N removal (from 46.0 to 54.5%) (J. Huang et al., 2000).

Biochar size and soil type also affect the WHC of biochar-amended soil matrix (Verheijen et al., 2019). For example, biochar particle size change can influence the WHC of soil by changing the pore space between particles (interpores) and by adding pores that were part of the biochar (intrapores) (Z. Liu et al., 2017). For example, in sandy soil amended with 2% (w/w) biochar (feedstock: mesquite; pyrolysis temperature: 400 ˚C), the decrease in biochar size reduced the WHC to almost 50% because grinding the biochar (<0.25 mm) resulted in the destruction of intrapores (Z. Liu et al., 2017). In another study, with 20% (v/v) biochar amendment (feedstock: mixed wood; pyrolysis temperature: 620 ˚C), the WHC in sandy soil increased from 43.1 to 53.3% as the biochar size increased from 0.05–1.00 to 2.0–4.0 mm (Verheijen et al., 2019).

3.1.4 | Microbial biomass enhancement

The overall microbial community structure and functional microbial species abundance may vary significantly in biochar-amended biofiltration systems for wastewater treatment (Sun et al., 2018). For example, the diversity of the microbial community, the relative abundance of nitrifying bacteria, and the activity of ammonia monooxygenase enzymes increased significantly in CWs containing 10–30% of biochar (v/v) compared with wetlands without biochar amendment (Liang et al., 2020). Biochar amendment could

also enhance the abundance of functional species involved in N transformations. *Nitrosomonas*, one of the dominant members of ammonium-oxidizing bacteria, accounted for ~0.01% of the overall microbial community in biochar-amended CWs, whereas its level in the control CW (i.e., without biochar amendment) was below the detection limit. *Nitrospira*, a common genus of nitrite-oxidizing bacteria, accounted for 0.12–0.34% of the overall microbial community in biochar-amended CWs, compared with 0.08% in the CW without biochar amendment (Liang et al., 2020). In a soil microcosm experiment conducted under denitrifying conditions, researchers found a significant increase in the diversity and transcript production of functional genes *nirK* (copper-based nitrite reductases) and *nosZ* (nitrous oxide reductase) in the biochar-amended soil (Harter et al., 2017). In a SWIS with biochar amendment, a similar increase was observed in the abundance of *nirS* (cytochrome-cd1 nitrite reductases), *nirK*, and *nosZ* (10^3, 10^2, and 10^3 times higher) compared with the conventional SWISs (Sun et al., 2018). The microbial abundance increase in the presence of biochar could be due to a variety of reasons. The larger surface area, greater pore space, and higher pH provided by biochar all favor biofilm formation. In addition, nutrients and minerals that are adsorbed to the biochar surface could be readily utilized by bacteria (Harter et al., 2017; Lehmann & Joseph, 2009).

3.2 | Biofiltration systems with biochar amendment for N removal

A limited number of studies have explored biochar application in wastewater treatment process for N removal (He, Ding, Wang, et al., 2018). In this review, we mainly focus on soil-based biofiltration systems with similar configurations (trickling filter, CW, and SWIS) that have incorporated biochar for N removal from on-site wastewater (Table 1). High-strength wastewater (150–2,852 mg N L^{-1}) has been applied to both unsaturated biochar (100%) trickling filters and saturated filters (Forbis-Stokes et al., 2018; Hunter & Deshusses, 2020; W. Li et al., 2016). For example, the NH_4^+ removal rate (0.075–0.100 kg N m^{-3} d^{-1}) was higher in the unsaturated biochar nitrifying column compared with the control gravel column (0.041–0.094 kg N m^{-3} d^{-1}) for a 1-yr operation (influent TN, 150–2,852 mg N L^{-1}) (Forbis-Stokes et al., 2018). In another trickling filter study, a 100% sand column was reported to remove about 8% of TN from domestic wastewater (38.5–63 mg N L^{-1}), and the addition of a subsequent 100% biochar filter module enhanced TN removal by 42% during an 18-mo experimental period (Tait et al., 2015). When woodchip biochar was amended to a sand filtration system (30% v/v) to treat dairy runoff (influent TN, 80 mg N L^{-1}), the effluent TN was <30 mg N L^{-1}, compared with 30–70 mg N L^{-1} in the sand-only biofilter effluent (Rahman et al., 2021). Constructed wetlands with biochar amendment (5–0% v/v) have been used for low-strength (TN <40 mg N L^{-1}) wastewater treatment, and effective TN removal (83.5–99.5% removal) has been reported (X. Chen et al., 2020; de Rozari et al., 2018; Liang et al., 2020; J. Xu et al., 2020). Constructed wetlands with 20% (w/w) biochar amendment showed enhanced TN and NH_4^+ removal (effluent TN, <0.6 mg N L^{-1}; NH_4^+–N, <0.04 mg N L^{-1}) compared with CW with 100% sand when treating secondary clarifies effluent (de Rozari et al., 2018). Similar observations of higher N removal during shorter experiments (35–90 d) were reported in other CW studies with biochar amendment compared with nonbiochar CWs (X. Chen et al., 2020; Guo et al., 2020; Liang et al., 2020; J. Xu et al., 2020; Zhou, Wang, et al., 2019; Zhou, Wu, et al., 2019). When influent N concentration increased from 67 to 160 mg N L^{-1}, the biochar-amended CW (50%, v/v) showed stable TN removal performance (47.1 vs. 41.3%), whereas the TN removal efficiency decreased to 19.2–22.1% in 100% gravel CW (Saeed et al., 2019). When biochar was amended to a SWIS, improved TN removal (30–80%) and slightly lower N_2O emissions (15–30 mg m^{-2} d^{-1}) were observed, compared with conventional SWIS without biochar addition (20–70% TN removal and 18–34 mg m^{-2} d^{-1} N_2O emission) (Qi et al., 2018).

At low temperatures, biochar-amended CWs also showed better performance compared with the conventional wetland (He et al., 2018b; He, Ding, Wang, et al., 2018). For example, during winter (4.9 ˚C), CWs with 10 and 20% (v/v) biochar amendment achieved 85.7 ± 6.2 and 91.7 ± 3.8% TN removal, which was statistically significantly higher than the conventional CW (73.6 ± 9.6%); this result was possibly due to the labile organic C released from biochar for denitrification (J. Li et al., 2019). Efficient TN removal (~50%) has also been demonstrated in a long-term experiment (125 d) using a 100% biochar–packed sequencing batch reactor treating synthetic wastewater at low temperature (5 ˚C) (He et al., 2018a).

3.3 | Factors affecting N removal by biochar-amended biofiltration systems

3.3.1 | Pyrolysis temperature

Different pyrolysis temperatures may affect biochar N removal capacity by changing (a) the biochar specific surface area (SSA) and (b) the types and abundance of the surface functional groups (Q. Yin et al., 2018; X. Zheng et al., 2018; L. Zhou et al., 2019). Contradictory results have been reported on the effect of pyrolysis temperature change on biochar adsorption of NH_4^+ and NO_3^-. The SSA of biochar increased significantly as the pyrolysis temperature increased (Rajkovich et al., 2011). For example, when pyrolysis temperature increased from 300 to 700 ˚C, the biochar SSA increased

TABLE 1 Summary of studies that applied biochar for N removal in biofiltration systems

Configuration	Biochar amendment ratio	Influent	Study scale (volume)	Biochar source	System performance	Reference
	%	mg N L^{-1}	L			
Trickling filter	100	150–2,852	laboratory (8.00)	pine pellets (900 °C[aa])	NH_4^+ removal rate 0.075–0.100 kg N m^{-3} d^{-1}	Forbis-Stokes et al. (2018)
Constructed wetland	50 (v/v)	67–160	laboratory (42.8)	–	41.3–47.1% TN removal efficiency	Saeed et al. (2019)
Constructed wetland	20 (v/v)	2.9–4.0, secondary clarifier effluent	mesocosm (240)	–	effluent TN <0.6 mg N L^{-1}; effluent NH_4^+ <0.04 mg N L^{-1}	de Rozari et al. (2018)
Constructed wetland	20 (v/v)	21.9 ± 1.17, synthetic wastewater	mesocosm (0.11)	peach pit (900 °C)	effluent TN <3.6 mg N L^{-1}	X. Chen et al. (2020)
Constructed wetland	0–30 (v/v)	39.4 ± 0.60, synthetic wastewater	laboratory (35.0)	bamboo (500 °C)	effluent TN <3.76 mg N L^{-1}; effluent NH_4^+ <1.4 mg N L^{-1}	Liang et al. (2020)
Constructed wetland	5 (w/w)	15.0, synthetic nitrified wastewater	laboratory (5.50)	–	effluent TN <1.0 mg N L^{-1}	J. Xu et al. (2020)
Constructed wetland	20 (v/v)	35.0–35.8, synthetic wastewater	laboratory (45.0)	cattail (300 °C)	45–52% TN removal efficiency; 38–55% NH_4^+ removal efficiency	Guo et al. (2020)
Subsurface wastewater infiltration system	10 (v/v)	38.5–43.7, septic tank effluent	laboratory (235)	corn cob (550 °C)	30–80% TN removal efficiency	Qi et al. (2018)
Subsurface wastewater infiltration system	100 (biochar filter after 100% sand filter)	38.5–63.0, domestic wastewater	laboratory (0.32)	green waste (550 °C)	42% improvement in TN removal	Tait et al. (2015)

Note. TN, total N.

[a]Pyrolysis temperature.

more than 10 times from 22.0 to 251.5 m^2 g^{-1}, and subsequently the NH_4^+ adsorption capacity increased from 2.0 to 5.9 mg N g^{-1} (X. Zheng et al., 2018). On the other hand, at high pyrolysis temperature (e.g., 700 ˚C), a decrease in H/C and (O+N)/C ratios on the biochar surface was observed, indicating the loss of polar functional groups on the biochar surface. This may lead to a reduced NH_4^+ adsorption capacity of the biochar (Yin et al., 2018). For example, a much higher NH_4^+ adsorption capacity (0.797 mg NH_4^+ N g^{-1} biochar) was observed in biochar produced at low pyrolysis temperature (300 ˚C) than in biochar produced at high pyrolysis temperature (700 ˚C) (0.130 mg NH_4^+ N g^{-1} biochar) (D. Xu et al., 2019). In another study, the adsorption of NH_4^+ decreased from 1.5–3.6 to 0.5–1.5 mg N g^{-1} when biochar pyrolysis temperature increased from 400 to 700 ˚C (Gai et al., 2014).

The increase of pyrolysis temperature had a slight positive effect on biochar adsorption of NO_3^- (Gai et al., 2014). For example, wood-based biochar produced at 800 ˚C had a higher nitrate adsorption capacity (0.2 mg NO_3^-–N g^{-1}) than biochar from the same source produced at 400 ˚C (0.05 mg NO_3^-–N g^{-1}) (Kameyama et al., 2016). Similarly, nitrate adsorption (0.1–0.2 mg NO_3^- N g^{-1}) was observed with biochar produced from digested sludge and sawdust at 700–800 ˚C, whereas no adsorption was observed in the biochar produced at lower pyrolysis temperatures (300–400 ˚C) (Kameyama et al., 2016; L. Zhou et al., 2019). In addition, no NO_3^- adsorption was observed by non–wood-based biochar, and the result was not affected by the biochar pyrolysis temperature change (400 vs. 800 ˚C) (Kameyama et al., 2016).

3.3.2 | Biochar source

The feedstock of biochar can be obtained from various sources and can be plant-based, manure-based, or agricultural/food processing residual–based biomass (Yuan et al., 2019). Biochar produced from woody materials (e.g., corn, oak, and pine) tends to have lower density (0.10–0.26 g cm^{-3}) compared with other sources, such as manure, food waste, paper waste, and poultry litter (0.12–0.65 g cm^{-3}) (Rajkovich et al., 2011). The adsorption capacity of NH_4^+ varies among biochar originating from different sources. For example, in the adsorption isotherm experiments, biochar produced from rice straw and sawdust showed higher NH_4^+ adsorption capacity (2.7–3.5 mg NH_4^+ N g^{-1}) than biochar produced from eggshells (1.8 mg NH_4^+ N g^{-1}) (D. Xu et al., 2019) and digested sludge (1.2 mg NH_4^+ N g^{-1}) (Tang et al., 2019). Lower NH_4^+ adsorption capacity in non–woody-source biochar may be due to the lower H/C ratio, which decreased the number of organic functional groups on the biochar surface (D. Xu et al., 2019).

3.3.3 | Amendment ratio and particle size

Constructed wetlands with various biochar volumetric amendment ratios (5, 10, 15, 20, and 25%) have been studied for N removal performance. The results showed that systems with >5% (v/v) biochar amendment had statistically higher TN and NH_4^+ removal compared with a control system without biochar amendment (de Rozari et al., 2018). Similar improved N removal rates were observed in other CW studies with >10% (v/v) biochar amendment ratios. For example, high NH_4^+ and TN removal efficiencies (49.7–63.5% NH_4^+ removal, 81.8–86.4% TN removal) were observed in a subsurface flow CW with 30% (v/v) biochar amendment (Deng et al., 2019). In another study, when 10–20% biochar (v/v) was amended to the sand layer of a CW, higher TN removal efficiency (90–93%) was observed compared with the CW without biochar addition (88%) (J. Li et al., 2019). Few clogging issues have been reported in the biochar-amended biofiltration systems, probably due to the low hydraulic loadings (20.5–49.2 L m^{-2} d^{-1}) applied in OWTSs (NYSDEC, 2014) compared with the trickling filters in wastewater treatment plants (2,600–293,000 L m^{-2} d^{-1}) (Elmitwalli et al., 2003; Logan et al., 1987). In addition, the porous structure of the filtration matrix was shown to result in better solute transport (e.g., nutrient transport) (Hosseini et al., 2020). On the other hand, pyrolysis reduces the mechanical strength of the source biomass; therefore, biochar is prone to breakage under compaction in the soil-based filtration systems. Typically, a 10–30% (v/v) biochar amendment ratio was applied to the filtration matrix to provide the mechanical strength needed (Ghavanloughajar et al., 2020).

4 | APPLICATION OF BIOCHAR IN BIOFILTRATION SYSTEMS FOR P REMOVAL

Untreated domestic wastewater contains 5–20 mg P L^{-1}, mainly from urine (470–1,070 mg P L^{-1}) and household detergents (Davis, 2011; IJC, 1971; Randall & Naidoo, 2018). The regulated P level in protected water bodies is 10 μg L^{-1} in the United States and 50 μg L^{-1} in Europe (Keeley et al., 2016). The excess discharge of P from domestic sewage may cause eutrophication in the aquatic system (Schindler et al., 2016). Furthermore, P is a nonrenewable resource, which makes P recovery from wastewater of significant importance (Cordell et al., 2011).

4.1 | P removal mechanisms

In municipal wastewater treatment plants, P is removed mainly by chemical precipitation and biological assimilation

(Rittmann & McCarty, 2001). Iron salts and aluminum sulfate are widely used for P precipitation, leading to higher sludge production for downstream treatment (Haas et al., 2000). On the other hand, P removal in soil-based OWTSs could be achieved via multiple mechanisms, such as adsorption; precipitation with inorganic minerals such as calcium (Ca), iron (Fe), aluminum (Al) ions; and biomass assimilation (Wehrmann et al., 2020). Phosphorus adsorption efficiency depends on sorbent characteristics, such as particle size, surface functional groups, and SSA (M. Li et al., 2016). Therefore, the filter media play an important role in P removal in soil-based OWTSs. The filter media that have been applied for P removal can be classified to three groups: (a) natural materials, such as gravel, peat, and limestone; (b) industrial byproducts, such as coal ash, fly ash, and oil shale; and (c) man-made products, such as lightweight expanded clay aggregate and synthesized clay-based filtration materials (e.g., Filtralite P) (Vohla et al., 2011). Recently, biochar has received more attention as an alternative filter media for P removal due to its low cost, wide availability, and low environmental effect (Yao et al., 2011). Phosphate removal by biochar could be achieved by (a) electrostatic attraction of negatively charged phosphate ion with positively charged biochar surface, (b) ion exchange, or (c) co-precipitation with Ca^{2+} and Mg^{2+} on the biochar surface (Bacelo et al., 2020; J. Liu et al., 2020; K. Xu et al., 2018). At low pH (<8.5), electrostatic adsorption was found to be the dominant removal mechanism (Park et al., 2018). At higher pH (>8.5), protons (H^+) are detached from the biochar surface, and the surface charge becomes more negative, leading to a reduction of the adsorption capacity of negatively charged pollutants and anions (R. Li et al., 2016).

4.2 | Biofiltration systems with biochar amendment for P removal

A limited number of studies have evaluated P removal performance by biochar-amended OWTSs (Table 2). Laboratory-scale column tests have been conducted to explore the biochar-amended CWs, trickling filters, and stormwater runoff bioretention systems for P removal, but very few field-scale studies have been conducted. Different biochar amendment approaches have been reported. In one approach, biochar was added to the top of the filtration system as a separate treatment layer. For example, biochar (0.2 m) was added on top of a sand layer (0.8 m) to treat domestic wastewater containing 1.9 mg P L^{-1}. An improved P removal efficiency (25.6%) was observed in the biochar-amended sand column compared with the 100% sand column (19.3%) when treating wastewater (Kholoma et al., 2016). In another study, the addition of a top biochar layer (0.2 m) to the sand layer (0.3 m) reduced the final effluent TP to 6.2 ± 1.0 mg P L^{-1}, compared with 7.6 ± 1.0 mg P L^{-1} in the control 0.5-m sand layer effluent when treating domestic wastewater (Kholoma et al., 2020). In another approach, biochar was mixed with the filter media. For example, 20% (w/w) biochar was amended to laboratory-scale CWs treating synthetic wastewater containing high P concentrations (36.1 mg P L^{-1}). Enhanced P removal (average effluent TP was 10.8 mg L^{-1}) was observed in the biochar-amended column, and the average effluent TP in the control gravel column was 15.0 mg P L^{-1} (Gupta et al., 2016). In yet another approach, a 100% biochar packed column was added as the treatment unit. A few case studies demonstrated that the 100% biochar–filled column was efficient in TP removal treating high-strength wastewater. For example, the corn cob biochar and wood biochar columns removed 71 and 83% of the influent P (37 mg P L^{-1}), respectively, which was much higher than the gravel wetlands (56% removal) when treating raw anaerobic digested effluent (Kizito, Lv, et al., 2017). In addition, an average 68% removal of TP was observed when the 100% biochar trickling filter was used to treat high-strength human waste (1,368 mg P L^{-1}) (Hunter & Deshusses, 2020; W. Li et al., 2016). A 100% biochar column has also been used to treat wastewater streams with low P concentrations. For example, in a pilot-scale study, the addition of a 100% biochar filter after a horizontal flow gravel wetland showed better phosphate removal in the long term (5 mo) when treating domestic wastewater with 0.3–9.9 mg P L^{-1}; 97% P removal was observed, compared with 87% in the control 100% gravel wetlands (Bolton et al., 2019). Another 100% biochar trickling filter was reported to remove up to 80% of TP when treating livestock wastewater (influent TP, 50 mg P L^{-1}) (Li et al., 2016c). The aluminum hydroxide–loaded biochar was reported to remove 8.3 g P kg^{-1} biochar when treating secondary clarifier effluent (0.9–6.4 mg P L^{-1}) (Zheng et al., 2019). A biochar-amended biofiltration system has also been tested as the P removal unit for simulated storm water runoff in a bioretention system. Bioretention systems are small, excavated areas that are backfilled with a mixture of highly permeable soil and organic matter in order to maximize infiltration and vegetation growth (Roy-Poirier et al., 2010). A 100% biochar packed bioretention system showed 47% P removal when treating low-P strength wastewater (<10 mg P L^{-1}) (Reddy et al., 2014).

On the other hand, biochar amendment may lead to a decreased P removal efficiency in the biofiltration system. In a mesocosm study, biochar was amended to CWs treating secondary clarifier effluent with low P concentrations (5.1–9.1 mg P L^{-1}). Total P removal efficiency was 70–100% in the control sand CW while it was lower at 40–60% in the CW with 25% (v/v) biochar amendment (de Rozari et al., 2016). When septage (22.5–26.3 mg P L^{-1}) was treated with both systems, a 30–50% TP removal efficiency was observed in the biochar-amended CW, whereas 70–80% TP removal was observed in the 100% sand CW (de Rozari et al., 2016). The decreased P removal efficiency may be due to the

TABLE 2 Summary of studies that applied biochar for phosphorus removal in biofiltration systems

Configuration	Biochar amendment ratio	Influent, P concentration	Reported P removal mechanism	Study scale	Biochar source	System performance	Reference
	%	mg P L^{-1}					
Constructed wetland	10–20 (w/w)	synthetic wastewater, 36.1	adsorption/ precipitation	laboratory	oak tree wood (600 °C[a])	62.3–70.1% TP removal	Gupta et al. (2016)
Constructed wetland	100	wastewater, 37	adsorption	laboratory	corn cob and wood biochar	71% (corn biochar) and 83% phosphate removal (wood biochar)	Kizito, Lv, et al. (2017)
Constructed wetland	25 (v/v)	secondary clarified wastewater, 5.1–9.1; septage, 22.5–26.3	adsorption	mesocosm	-	40–60% TP removal (clarifier effluent), 30–50% TP removal (septage)	de Rozari et al. (2016)
Fixed-bed column	100	secondary treated municipal clarifier effluent, 0.9–6.4	adsorption	laboratory	biochar/AlO_2H composite (600 °C)	8,346 mg P kg^{-1} biochar	Y. Zheng et al. (2019)
Trickling filter	100	synthetic wastewater, 50	adsorption	laboratory	palm residues (700 °C)	68% phosphate removal	W. Li et al. (2016)
Nitrification trickling filter	100	high-strength human wastewater, 1,368	adsorption	laboratory	pine	89% phosphate removal	Hunter et al. (2020)
Bioretention system	100	simulated urban storm water runoff, 0.5–1	adsorption	laboratory	waste wood (520 °C)	47% TP removal	Reddy et al. (2014)
Fortified filter beds	0.2-m biochar layer on top of 0.8 m sand	pretreated domestic wastewater, 4.5–8.0	adsorption	laboratory	–	25.6% phosphate removal	Ezekiel Kholoma et al. (2016)
Packed bed bioreactor	0.2-m biochar layer on top of 0.3 m sand	raw household wastewater, 10.0 ± 1.8	adsorption	laboratory	wood chips (500 °C)	18.4% phosphate removal	Ezekiel Kholoma et al. (2020)

Note. TP, total P.
[a]Pyrolysis temperature.

negative surface charge of the selected biochar and the competition from other anions for exchange sites on the biochar surface (de Rozari et al., 2016; Yao et al., 2012).

4.3 | Factors affecting P removal by biochar-amended biofiltration systems

Adsorption was the main P removal mechanism in CWs, and P removal efficiency may decrease in the long term due to the saturation of adsorption sites (Y. Gao et al., 2018). However, the observed adsorption in N-removing biofilters was less compared with mineral precipitation and recrystallization reactions, which showed the importance of different mechanisms on P removal (Wehrmann et al., 2020). Studies have shown the overall surface charge of biochar was negative, resulting in the limited anionic pollutants adsorption capacity (B. Chen et al., 2011). In this section, factors that may affect biochar P removal efficiency are investigated, and innovative solutions to enhance P removal efficiency are summarized.

4.3.1 | Pyrolysis temperature

Contradictory results have been reported on the effect of pyrolysis temperature change on biochar P adsorption capacity. The increase of pyrolysis temperature increased the inorganic elements (e.g., Mg, Na, Mn, Fe, Al, and Ca) content in the biochar, which enhanced the extent of phosphate reaction with metal ions and subsequently increased the precipitation (Figure 2) (Blanco et al., 2016). For example, the phosphate adsorption capacity of crawfish biochar increased about eight times (from 9.5 to 70.9 mg P g^{-1}) when the pyrolysis temperature increased from 200 to 800 °C (Park et al., 2018). On the other hand, the P removal efficiency by macro-algae decreased from 18.2 to 13.0 mg P g^{-1} when the pyrolysis

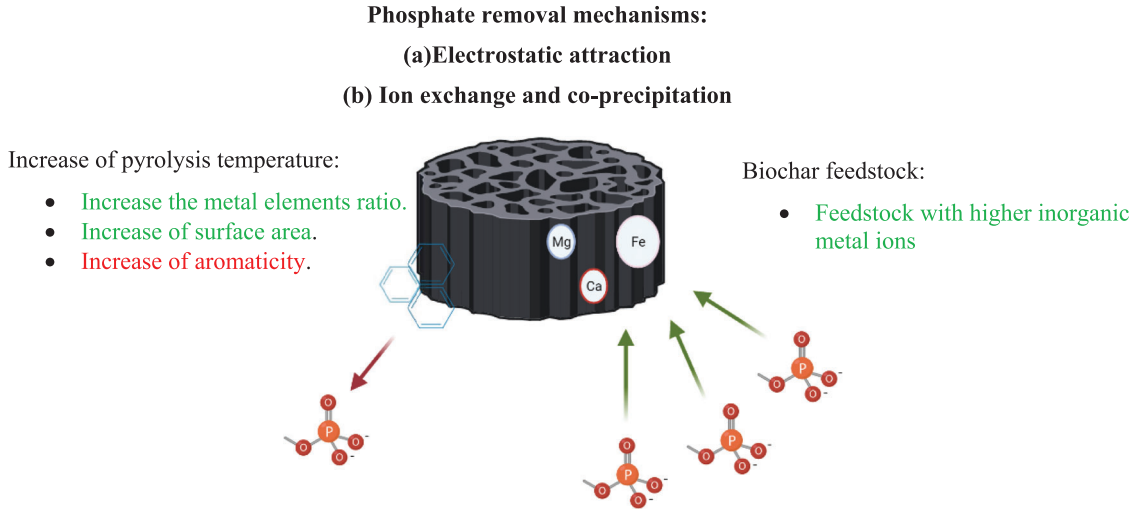

FIGURE 2 Factors that may affect P removal efficiency. Green shows positive effect on phosphate removal; red shows negative effect

temperature decreased from 800 to 400 °C. This was mainly due to the decrease of biochar surface polarity and the increase in aromaticity of the biochar, which led to a reduction of adsorption through anion exchange (Figure 2) (Jung et al., 2016).

4.3.2 | Biochar type and modification

The P adsorption capacity varies significantly with biochar generated from different sources. Biochar generated from wood, rice husk, sawdust, corncobs, sugar cane, peanut shells, and plant waste material showed a maximum P adsorption capacity of 1.6–13.2 mg P g^{-1} without any surface modification (Jung et al., 2015; Kizito, Luo, et al., 2017; Y. Li et al., 2019; Trazzi et al., 2016). On the other hand, biochar produced from sewage sludge with higher cation content (e.g., Ca, Al, Fe, and Mg) showed a greater potential of phosphate adsorption (49.9 mg PO_4^{3-} g^{-1}) compared with biochar produced from walnut shells (0.7 mg PO_4^{3-} g^{-1}) in single-solute adsorption tests (Q. Yin et al., 2019). If the solution pH is lower than the pH_{pzc}, the biochar surface is positively charged, and the biochar is more effective in phosphate removal (Jiang et al., 2019). For example, the pH_{pzc} of Mg-loaded biochar increased to 10.1–10.5; subsequently, the TP removal increased from 6.8 to 31.2 mg g^{-1} (Jiang et al., 2019). In another study, the pH_{pzc} of Mg-modified biochar increased from 5.6 to 8.2, which resulted in an increase in CEC from 1.6 to 31.6 mg g^{-1} (Xiao et al., 2020).

Metal oxides and hydroxides have been used for biochar surface modification to enhance the P removal capacity due to their high abundance, low cost, environmental friendliness, and chemical stability (M. Li et al., 2016). More efficient anion pollutant removal has been observed by functionalizing the biochar surface with metal oxide functional groups, such as aluminum hydroxide oxide (AlOOH) (Zhang & Gao, 2013). Aluminum-, Fe-, and Mg-impregnated biochar has been tested for phosphate adsorption, and significantly higher adsorption capacity was achieved (46.6–887 mg P g^{-1}) compared with unmodified biochar (8.1–12.0 mg P g^{-1}) (Bacelo et al., 2020; Fang et al., 2014; Jiang et al., 2019; Michalekova-Richveisova et al., 2017; Novais et al., 2018; Peng et al., 2019; Vikrant et al., 2018; K. Xu et al., 2018; Yao et al., 2013). Double-layered aluminum hydroxide has been used to coat the biochar surface; the resulting enhanced P adsorption capacity (410 mg P g^{-1}) was mainly due to the increased anion sorption capacity on the biochar surface (Muisa et al., 2020). In recent studies, biochar with chitosan, quaternary ammonium salt, and lanthanum [$La(NO_3)_3{\cdot}6H_2O$] modification was reported to achieve 209 mg P g^{-1} removal in an adsorption experiment (Y. Huang et al., 2020). Adsorption capacity was not significantly affected by temperature; therefore, the modified biochar was suggested to be used for phosphate removal from onsite wastewater (Y. Huang et al., 2020).

4.4 | P recovery from wastewater by biochar

Phosphate removal in wastewater treatment plants was mainly achieved by struvite ($NH_4MgPO_4{\cdot}6H_2O$) precipitation (Hunter & Deshusses, 2020). However, the solubility of struvite was low (169.2 ± 4.3 mg L^{-1} at 25 °C), and the precipitated P was not readily available for plant uptake (Bhuiyan et al., 2010; Maroušek et al., 2020). On the other hand, P that has been removed by the biochar-amended filtration systems can be slowly leached out, and the biochar has the potential to be applied as a fertilizer amendment (Vikrant et al., 2018). The desorbed P was reported to be related to the phosphate concentration gradient in the solution and to biochar surface characteristics. Therefore, the desorption rate increased

when the initial adsorbed phosphate level was higher (Trazzi et al., 2016). When biochar was added to agricultural soil, the P availability for plants increased by a factor of 4.6, independent from the biochar source or pyrolysis temperature (Glaser & Lehr, 2019). A case study has demonstrated that the level of phosphate desorbed from a biochar-amended sand filter (a 0.2 m layer of biochar on the top of 0.8 m of sand) was two times higher than that desorbed from the control sand filter (31.3 ± 3.4 mg P g^{-1}) (Kholoma et al., 2019). In another study, P-enriched biochar was readily available for plant uptake because the level of P extracted from biochar was 7.6 mg P g^{-1}, which was higher than the optimum P level (0.05 mg P g^{-1} soil) needed for plant growth (Yao et al., 2013). Engineered biochar with aluminum hydroxide [$Al(OH)_3$] surface modification removed 8.3 mg P g^{-1} of biochar from secondary treatment effluent, and this biochar was able to promote mung bean [*Vigna radiata* (L.) R. Wilczek] growth as an agricultural fertilizer (Y. Zheng et al., 2019). Magnesium was also considered as an appropriate cation for biochar surface modification because Mg was involved in the chlorophyll formation of plants (Fang et al., 2014; J. Liu et al., 2020). It was reported that phosphate could be adsorbed to Mg-enriched biochar up to 100 mg P g^{-1} and was also mostly bioavailable for plant growth (Yao et al., 2013). The potential use of MgO-impregnated magnetic biochar as a phosphate-based fertilizer showed significantly enhanced ryegrass (Lolium perenne L.) growth in height and weight (R. Li et al., 2016). The Ca- and Mg-modified biochar consistently released small amounts of adsorbed P (1.4–1.7%) when fresh distilled water was used for extraction (Fang et al., 2015), indicating that biochar could be used for long-term P recovery. Similar levels of P release (0.7–1.5%) were reported when Mg-modified biochar was used for P recovery (Nardis et al., 2020). The slow release of P (i.e., desorption reached equilibrium in 40–50 h) was preferred because it allowed P to be readily available for plant uptake before it was leached to the groundwater (Haddad et al., 2018).

Although nutrient-enriched biochar can serve as an effective soil amendment, there are concerns that other contaminants may be leached from biochar during soil application. For example, heavy metals may be leached from biochar during the P recovery process. However, it was shown that calcium hydroxide [$Ca(OH)_2$] addition before pyrolysis could immobilize the metals in biochar and reduce metal release when biochar was applied to soil (100 and 70 times less for copper and zinc, respectively) (Antunes et al., 2018). Biochar source also has a great effect on the levels of heavy metals that might be leached out. For example, biosolids tend to have higher levels of heavy metals when they were used for biochar production (Shakoor et al., 2021). Polycyclic aromatic hydrocarbons and other organic pollutants could also be leached out from biochar (Shakoor et al., 2021). It has been suggested that slow pyrolysis (a few seconds for fast pyrolysis vs. hours for slow pyrolysis) at temperatures higher than 700 °C would decrease polycyclic aromatic hydrocarbon levels and immobilize organic pollutants in biochar (Bair et al., 2016; Fabbri et al., 2013).

5 | APPLICATION OF BIOCHAR IN BIOFILTRATION SYSTEMS FOR PATHOGEN REMOVAL

Pathogens that are present in domestic wastewater can pose a great health risk if not properly treated (USEPA, 2003). Bacterial pathogens (e.g., *Escherichia coli*, *Legionella pneumophila*, *Leptospira* spp., *Salmonella* spp., *Shigella* spp., *Vibro cholera*, and *Yersinia enterocoliticia*) and viral pathogens (e.g., adenovirus, enterovirus, hepatitis A, norovirus, reovirus, rotavirus, and echovirus) have been found in onsite domestic wastewater effluent (Lusk et al., 2017). Conventional OWTSs can remove only 24–83% of pathogens, and viral particles in the effluent can travel long distances in groundwater (DeBorde et al., 1998; Olson et al., 2005). For example, in a case study, water samples were collected from various coastal areas in Florida and were tested for fecal coliform indicators and human enteric pathogens (e.g., *Salmonella* and *Shigella*). The results demonstrated that areas with highest risk of pathogen pollution were close to OWTSs. Advanced OWTSs, such as innovative drain fields and biofilters, could remove up to 99.9% of *E. coli* using different adsorption media, including tire crumbs and biochar (Chang et al., 2010; Kaetzl et al., 2019).

5.1 | Pathogen removal mechanisms

Pathogen removal by a filtration-based OWTS depends on (a) the efficiency of physical entrapment in the pores of filtration media and (b) the adsorption capacity of the filtration media (Perez-Mercado et al., 2019; M. Wang et al., 2021). Adsorption was reported to be one of the dominant pathogen removal mechanisms in OWTS studies (Gwenzi et al., 2017; Perez-Mercado et al., 2019; M. Wang et al., 2021). Biochar has been reported to be effective in fecal coliform removal due to its high adsorption capacity resulting from its high surface area compared with sand particles (143 m^2 g^{-1} for biochar vs. <0.004 m^2 g^{-1} for sand) (Kaetzl et al., 2019). When the biochar surface was loaded with a zerovalent iron (ZVI)–silver (Ag) nanoparticles (NPs) complex, the growth of *E. coli* was completely inhibited, compared with unmodified biochar (4×10^8 colony-forming units ml^{-1} *E. coli*) (Zhou et al., 2014). Silver nanoparticles (Ag-NPs) are known as antimicrobial particles due to a variety of mechanisms, such as disruption of cell wall and cytoplasmic membrane, denaturation of ribosomes, interruption of ATP production, and

interference of DNA replication (I. X. Yin et al., 2020). The superior pathogen inhibition performance suggested that Ag-NP–modified biochar could be applied for pathogen removal (Zhou et al., 2014). In addition, other metal nanoparticles with high surface-to-volume ratio increase the production of reactive oxygen species (e.g., free radicals) and cause cell membrane disruption (Thukkaram et al., 2014). For example, magnetic biochar (with iron oxide precipitation on the surface) was reported to have antibacterial function and removed almost 100% of *E. coli* and *Staphylococcus aureus* in batch experiments (with 200 mg L^{-1} biochar) (Fu et al., 2020).

5.2 | Pathogen removal performance in biochar-amended biofiltration systems

Most of the limited number of studies that reported the effect of biochar amendment on pathogen removal from domestic wastewater (Gwenzi et al., 2017) focused on *E. coli* removal (Boehm et al., 2020). Anaerobic biofiltration of raw wastewater using biochar showed substantially higher removal of *E. coli* (99.5%), enterococci (99.6%), and bacteriophages (98.6%) than sand filters (Kaetzl et al., 2019). However, the specific pathogen removal efficiency may depend on the type of pathogen of interest due to the differences of their isoelectric points (i.e., the pH at which a molecule is electrically neutral) and the hydrophobicity of the pathogen (Perez-Mercado et al., 2019). On the other hand, very few studies have evaluated viral pathogen removal efficiency in biochar-amended biofiltration systems. For example, a 100% biochar filter removed 2.7 $\log_{10}$ *Salmonella* spp., whereas only 1.0 $\log_{10}$ of *Escherichia* virus MS2 was removed when treating contaminated groundwater (Sidibe, 2014). In stormwater biofilters, 30% (v/v) biochar amendment resulted in 3.9 and 1.8 $\log_{10}$ removal of pathogenic *E. coli*, and *Escherichia* virus MS2, respectively, whereas only 0.3 $\log_{10}$ removal of pathogenic *E. coli* was observed in sand biofilters (Afrooz et al., 2018).

Contradictory results have been reported in biochar-amended filtration systems on pathogen removal performance in response to hydraulic loading changes. For example, in a biochar-amended (33%, v/v) stormwater biofilter, an additional 0.5–1.0 $\log_{10}$ *E. coli* removal was achieved at higher hydraulic loading compared with the control (sand only) system (sand pore water velocity 22.4–25.4 cm h^{-1} vs. biochar-amended biofilter pore water velocity 20.0–21.7 cm h^{-1}) (Kranner et al., 2019). On the other hand, the hydraulic loading rate increase (from 23 to 39 L m^{-2} d^{-1}) did not affect *E. coli* removal (2.48–3.39 $\log_{10}$) in the combined vertical–horizontal flow biochar filtration system (Dalahmeh et al., 2019).

Biochar type is another important factor influencing pathogen removal performance in biochar-amended OWTSs (Gwenzi et al., 2017). Previous studies have demonstrated that wood-based biochar maintains the plant cell structure and contains interconnected pores with 5–10 μm diameter, which are optimal for bacterial retention and entrapment, although non–wood-based biochar was more amorphous, with larger interconnected pores (up to 300 μm in diameter) (Abit et al., 2012). Biochar originated from forestry wood waste (pyrolysis temperature 700 ˚C) was reported to remove 92–99% *E. coli* from synthetic storm water (Lau et al., 2017). Biochar has a more negative surface charge at higher pH (>8) (R. Li et al., 2016). At high pH (>8), the zeta potential of both oxidized and unoxidized biochar was reduced by 40–60 mV (compared with the zeta potential at pH 2), which was similar to the zeta potential reduction of *E. coli* (a reduction of 80 mV) (Suliman et al., 2017). Therefore, the pathogen removal efficiency in biochar-amended OWTSs may decrease significantly at high pH due to the electrostatic repulsion between biochar and bacteria.

6 | APPLICATION OF BIOCHAR IN BIOFILTRATION SYSTEMS FOR PPCP REMOVAL

Trace organic compounds, such as PPCPs, were frequently detected in domestic wastewater (Lusk et al., 2017). The effect of PPCPs on the aquatic environment was concerning due to the possibility of continuous harm on aquatic organisms (Benotti et al., 2009; Daughton & Ternes, 1999; Quesada et al., 2019). A variety of pharmaceuticals have been detected in urine samples, septic systems, and the surrounding environment (Q. Gao et al., 2019; Godfrey et al., 2007; Lienert et al., 2007). Therefore, it is important to enhance PPCP removal in OWTSs, and biochar has been investigated for its ability to remove various PPCPs (de Andrade et al., 2018; Inyang & Dickenson, 2015).

6.1 | PPCP removal in biochar-amended biofilters and the removal mechanisms

Various PPCPs have been detected in STE in a limited number of case studies (Y. Y. Yang et al., 2016). Based on data collected from two different studies in North Carolina, USA, and Denmark, the most frequently detected PPCPs (>60% of the tested samples) in STE were ibuprofen ($C_{13}H_{22}O_3$), caffeine ($C_8H_{10}N_4O_2$), homosalate ($C_{16}H_{22}O_3$), salicylic acid ($C_7H_6O_3$), naproxen ($C_{14}H_{13}NaO_3$), methyl dihydrojasmonate ($C_{13}H_{22}O_3$), and hydrocinnamic acid ($C_9H_{10}O_2$) (Del Rosario et al., 2014; Matamoros et al., 2009). Among the detected PPCPs, the chemicals with the highest concentrations detected in OWTSs were ibuprofen (73.8 μg L^{-1}), caffein (70.8 μg L^{-1}), and salicylic acid (67.7 μg L^{-1}) (Del

Rosario et al., 2014; Matamoros et al., 2009). Physical, chemical, and biological removal mechanisms have been proposed in biochar-amended CWs when treating domestic wastewater containing PPCPs: (a) adsorption of PPCPs to the biochar surface via van der Waals forces and hydrogen bonds with functional groups, electrostatic interaction, ion exchange, and surface complexation; (b) absorption (hydrophobic partitioning); (c) biodegradation by microorganisms at biochar surface; and (d) pore filling and intraparticle diffusion (Cheng et al., 2021; L. Li et al., 2019; Y. Li et al., 2014).

Both 100% biochar filtration unit and biochar-amended filtration systems have been tested to enhance PPCP removal efficiency. In a bench-scale study, a 100% biochar filter and a 100% sand filter were compared when treating wastewater spiked with selected PPCPs (carbamazepine, metoprolol, ranitidine, and caffeine). Adsorption was found to be the main removal mechanism for carbamazepine and metoprolol (95–99% removal) in the biochar column. Ranitidine and caffeine were either adsorbed or biologically degraded up to 99% in the biochar column. On the other hand, the sand column showed lower removal (36–73%) for the tested PPCPs, except for ranitidine (96–99%) (Dalahmeh et al., 2018). Biochar has also been incorporated to the filtration system as a filtration layer to investigate its PPCP removal performance. A sand filter with 10 cm of biochar on the top (13% v/v) and 30 cm of granular activated carbon (C) at the bottom (43% v/v) was used for long-term post-treatment of wastewater treatment plant effluent. This biochar-amended system was effective in removing organic micropollutants (i.e., anti-epileptic drugs gabapentin and carbamazepine, the anti-inflammatory drug diclofenac, and the antibiotic clarithromycin) (Brunsch et al., 2018).

There is a great potential to amend biochar to biofiltration system for enhanced PPCP adsorption (Cheng et al., 2021). When biochar was amended to the soil matrix (0.5% w/w), the partitioning and removal of selected pharmaceuticals (carbamazepine and propranolol) increased by threefold (Williams et al., 2015). Biochar was also amended to different filtration matrices to enhance PPCP removal. In a case study, PPCP removal performance was compared among different filtration matrices: (a) 100% silica sand, (b) 100% ZVI, (c) biochar-amended sand (50% v/v), and (d) 10% ZVI + 40% biochar + 50% sand (v/v). A variety of PPCPs (i.e., carbamazepine, caffeine, sulfamethoxazole, 3,4-methylenedioxyamphetamine, 3,4-methylenedioxymethamphetamine, ibuprofen, gemfibrozil, and naproxen) were selected for the column tests (Y. Liu et al., 2019). The column with a combination of ZVI, biochar, and sand showed the highest PPCP removal efficiency (>97% removal of all PPCPs at 10 μg L^{-1} of each chemical) within the top 10 cm of the column. The column with 50% biochar was also effective in PPCP removal; however, the removal was completed at a column depth of 20–30 cm. The 100% sand control column did not show effective removal for the tested PPCPs except sulfamethoxazole (0.3 μg L^{-1} in the effluent) (Y. Liu et al., 2019).

6.2 | Factors affecting PPCP removal by biochar-amended biofiltration systems

6.2.1 | Pyrolysis temperature

Biochar produced at different pyrolysis temperatures may remove PPCPs via different adsorption mechanisms (Oh & Seo, 2016; Rajapaksha et al., 2019). At low pyrolysis temperature (250–450 ˚C), more abundant organic functional groups are present on the biochar surface, and increased partitioning (due to noncarbonized organic residuals) could increase PPCP removal (Oh & Seo, 2016). At high pyrolysis temperature (550–900 ˚C), increased aromaticity and larger surface area were the dominant factors contributing to PPCP removal by biochar (Oh & Seo, 2016; Rajapaksha et al., 2019). Therefore, biochar obtained from the same source at various pyrolysis temperatures may have significantly different adsorption capacity of the selected PPCP. For example, in batch adsorption tests, rice straw biochar obtained at high pyrolysis temperature (700 ˚C) showed two to six times higher adsorption of oxytetracycline and tetracycline (18 mg g^{-1} for oxytetracycline and 8.0 mg g^{-1} for tetracycline) than the adsorption levels observed with biochar produced at low pyrolysis temperature (300 ˚C) (M. Li et al., 2017; H. Wang et al., 2017). On the contrary, a higher adsorption capacity of triclosan and 2,4-dichlorophenol (40–50 mg g^{-1}) was observed in rice straw biochar produced at low pyrolysis temperature (250 and 400 ˚C), whereas <30 mg g^{-1} adsorption of both compounds was observed in biochar produced at high pyrolysis temperature (550–900 ˚C) (Oh & Seo, 2016).

6.2.2 | Biochar surface modification

Two types of biochar modification methods have been successfully applied for enhanced PPCP removal: (a) biochar with acid/alkali treatment to enhance the abundance of oxygenated functional groups on its surface (M. B. Ahmed et al., 2016; L. Li et al., 2019; Sophia & Lima, 2018) and (b) biochar composites impregnated with nanomaterials (e.g., Fe and Ag) (Sophia & Lima, 2018). For acid/alkali treatment, potassium hydroxide (KOH) was commonly used for tetracycline removal. In batch adsorption tests, up to 58.8 mg g^{-1} of tetracycline adsorption was observed in the KOH-modified biochar, compared with 16.9 mg g^{-1} in the unmodified biochar. The enhanced adsorption capacity was mainly due to the higher surface area as well as more oxygenated functional groups on the biochar surface (P. Liu et al., 2012). Nanoma-

terials have also been impregnated in biochar composite to enhance PPCP removal. For example, the adsorption capacities of a selected group of nonsteroidal anti-inflammatory drugs (salicylic acid [683 mg g^{-1}], naproxen [533 mg g^{-1}], and ketoprofen [444 mg g^{-1}]) were 11–45 times higher in iron oxide (Fe_2O_3)-impregnated biochar composites compared with the levels reported in unmodified biochar and activated C (Ahmed & Hameed, 2018; Anfar et al., 2020; Baccar et al., 2012; Karunanayake et al., 2017; Song et al., 2017). The higher adsorption capacities of caffeine, ibuprofen, and acetylsalicylic acid were also observed in the iron oxide nanoparticle–impregnated biochar (Liyanage et al., 2020). In addition, enhanced PPCP removal has been observed in biochar with both alkaline treatment and nanoparticle impregnation. For example, sodium hydroxide (NaOH) was added to Fe^{2+}/Fe^{3+} ion solution, and then the biochar was treated with the prepared solution through a co-precipitation process. The produced magnetic chitosan biochar removed 96.4% of diclofenac, 98.8% of ibuprofen, and 95.2% of naproxen in batch adsorption tests using synthetic aqueous solutions (Mojiri et al., 2019).

6.2.3 | Environmental pH

The pH effect on the PPCP adsorption capacity of biochar depends on the pK_a of the target PPCP and the pH_{pzc} of the biochar. The maximum adsorption of a specific PPCP is achieved when the minimum repulsion between the compound and the biochar surface occurs (pK_a of a specific compound < solution pH < pH_{pzc} of biochar) (Liyanage et al., 2020). In general, acidic pH favors PPCP removal due to the lack of deprotonation from the biochar surface at low pH (Oh & Seo, 2016). For example, the adsorption of triclosan and ibuprofen by biochar was up to 500 mg g^{-1} at low pH (4–7), compared with 4 mg g^{-1} at higher pH (>10). In another case study, the maximum adsorption of sulfonamide and sulfamethazine on biochar was observed at low pH (3–4.5) (Peiris et al., 2017).

6.2.4 | Wastewater composition

The complex composition of wastewater could significantly affect the removal process of PPCPs by biochar, mainly due to the competition for adsorption sites (Rajapaksha et al., 2019). For example, the elevated levels of phosphate in wastewater may decrease triclosan adsorption to biochar due to the competition for adsorption sites because both phosphate and triclosan are in anionic form in wastewater (Kimbell et al., 2018). On the other hand, the adsorption of heavy metals such as Cd^{2+} was reported to increase the positive charge on biochar surface, thus increasing the adsorption of sulfamethoxazole to biochar (Han et al., 2013). High salinity and high ionic strength (e.g., electrolyte ions Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^-, and SO_4^{2-}) in wastewater have been reported to improve ibuprofen and sulfamethoxazole adsorption to biochar, whereas elevated levels of carbonates and humic acid had a negative effect on pharmaceutical adsorption due to the competition for active adsorption sites (Lin et al., 2017).

7 | FUTURE PERSPECTIVES

Although studies have been conducted on biochar application in contaminant removal from wastewater, most studies have focused on laboratory-scale biochar-amended biofiltration systems. To date, there is no comprehensive understanding of the role that biochar plays in soil-based OWTSs. For example, contradictory results have been reported on how biochar preparation methods affect the biochar-amended biofiltration system performance in N, P, and PPCP removal (Figure 3). The biochar amendment ratio, the feedstock C and N content, and the incubation period can all affect the N and P transformations in the biofiltration system. Specifically, few studies have focused on the long-term performance of biochar-amended biofiltration systems (e.g., the effect of biochar aging) in nutrient removal. The long-term change in the biochar surface organic functional groups, its adsorption ability, and the microbial community structure on the biochar surface may affect the dominant contaminant removal mechanism that biochar provides during the wastewater treatment process. In addition, the biochar-amended biofiltration system response to changes in operation conditions (e.g., hydraulic loading and wastewater composition) and changes in environmental conditions (e.g., temperature) has not been systematically evaluated. Moreover, biochar amendment to OWTS may improve pathogen removal performance due to adsorption and entrapment of bacteria, whereas virus removal has not been systematically investigated. Engineered biochar with surface modification has been demonstrated to be effective in P recovery, and the subsequent biochar could be reused as a soil fertilizer. These studies suggest that the slow release of P from modified biochar can prevent overleaching of P to groundwater. In addition, biochar surface modification may facilitate the immobilization of contaminants (e.g., heavy metals, PPCPs, and pathogens). The regeneration of biochar has been a focus of biochar use in the municipal wastewater treatment process (Cheng et al., 2021); however, it is usually not a feasible option for OWTSs. Because biochar adsorbs not only nutrients but also other contaminants (e.g., pathogens and PPCPs), it might be a drawback if the biochar is re-applied in agriculture, with the potential leaching of these contaminants. The development of stan-

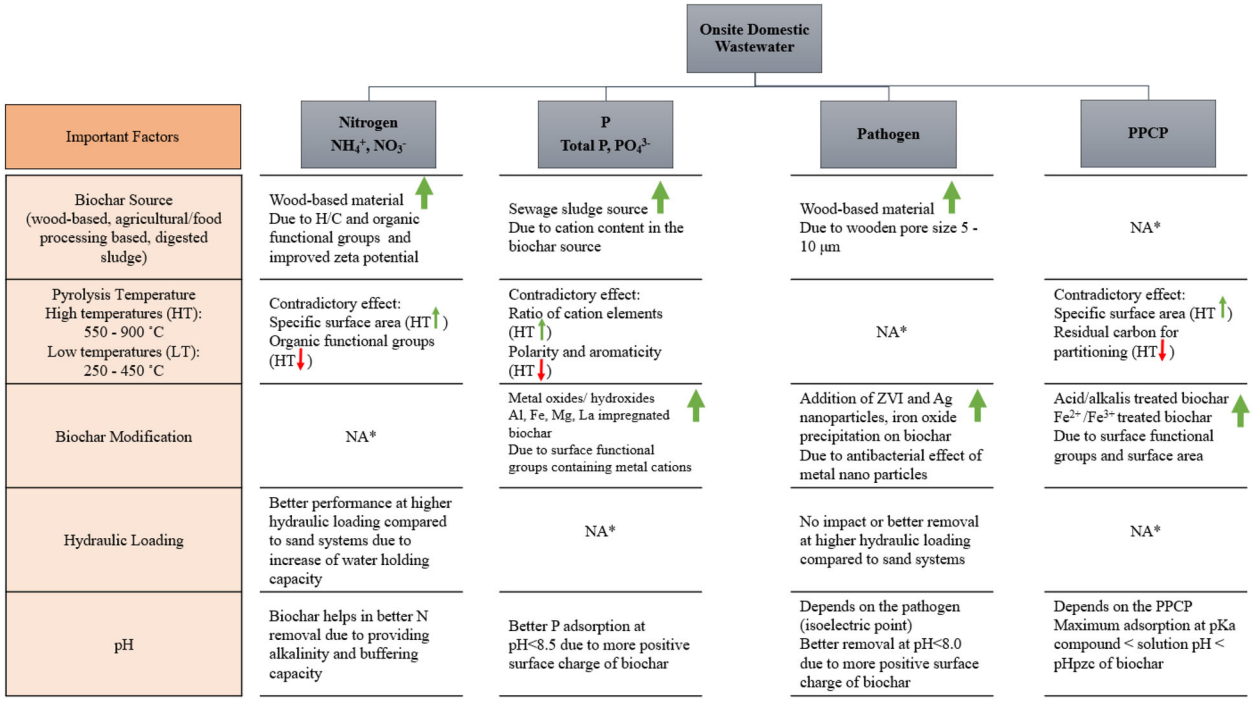

FIGURE 3 Summary of the most important factors that affect the performance of biochar-amended onsite wastewater treatment systems in removing various contaminants from domestic wastewater. Green arrows show positive effects on each pollutant removal; red arrows show negative effect. *Not enough information about that parameter to be included in this chart. PPCP, pharmaceuticals and personal care product; ZVI, zerovalent iron

dardized techniques for engineered biochar production, its integration to the OWTSs, and tracking uptake and release of P and other contaminants in field trials will open new avenues for research in this direction.

8 | CONCLUSION

Biochar has been amended to trickling filters, CWs, and subsurface wastewater infiltration systems to treat onsite wastewater. Enhanced N removal was achieved through a combination of various mechanisms, including adsorption, alkalinity supplement, and increased water holding capacity. Biochar can also serve as a growth medium to enhance the abundance of functional species involved in N transformation. Phosphorus removal by biochar-amended biofiltration systems was mainly through adsorption and precipitation, and PPCP removal was achieved via a combination of adsorption, absorption, and biodegradation. Biochar surface modification significantly enhanced the removal capacity of both P and PPCP by increasing the positive surface charge via addition of metal oxide functional groups. Pyrolysis temperature change, biochar source, biochar particle size, and amendment ratio all affected N, P, and PPCP removal performance. Specifically, contradictory results of N, P, and PPCP removal performance in biochar-amended biofiltration systems have been observed at increased pyrolysis temperature because higher pyrolysis temperature increases the surface area and decreases the abundance of oxygenated functional groups. Operational conditions, such as hydraulic loading, wastewater composition, and environmental conditions (e.g., pH), also have significant effects on the performance of biochar-amended biofiltration systems in removing nutrients, PPCPs, and pathogens. Biochar-amended biofilters showed better N removal at low temperatures and less N_2O emission. Biochar also holds great potential to recover P from onsite wastewater and can be applied as soil fertilizer.

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AUTHOR CONTRIBUTIONS

Zahra Maleki Shahraki: Data curation; Formal analysis; Investigation; Writing – original draft. Xinwei Mao: Conceptualization; Funding acquisition; Project administration; Resources; Supervision; Validation; Writing – review & editing.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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