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Exploring the engineering-scale potential of designer biochar pellets for phosphorus loss reduction from tile-drained agroecosystems

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ABSTRACT

Artificial drainage has led to significant amounts of non-point dissolved reactive phosphorus (DRP) loss from tile-drained agroecosystems, jeopardizing water quality and triggering harmful algal blooms. Designer biochar has shown great promise on the laboratory scale for removing DRP from contaminated water. However, whether its removal performance, stability, and engineering value can be sustained under field conditions over time remains unclear. This study reported the first engineering application of designer biochar pellets used in an intensely tile-drained agroecosystem to reduce DRP losses from drainage water. Two types of designer biochar pellets with different particle sizes (Phase I - biochar pellets size 2-3 cm vs. Phase II - biochar pellets size <1 cm) were manufactured and placed into the specifically designed phosphorus removal structure (i.e., biochar-sorption chamber) to capture DRP from tile drainage water. Field demonstrations revealed that small-sized biochar pellets (<1 cm) were significantly more efficient at capturing DRP than larger pellets (2-3 cm). A comprehensive analysis further indicated that multi-factors could affect the performance of designer biochar pellets in DRP loss reduction, such as influent DRP concentrations, drainage flows, and biochar pellet sizes. Techno-economic analysis and life cycle assessment indicated that the designer biochar pellets have notable economic and environmental benefits. On the pilot scale, the average production cost of designer biochar pellets was $413/ton biochar. The average DRP removal cost was $359±177/kg DRP for tile-drained agroecosystems under wide economic and system design parameters. Furthermore, utilization of designer biochar pellets to remove DRP from drainage in combination with subsequently using spent biochar as a soil amendment provides environmental benefits to achieve negative global warming potential (-200 to -12 kg CO_2 eq/kg DRP removal) and energy production. Overall, this work offers a novel strategy to explore the potential for engineering-scale application of biochar for sustainable water quality protection and helps elucidate the costs and benefits in the context of watershed nutrient loss management.

1. Introduction

Although agricultural intensification boosts crop production to meet food demands, it is a major driver of global water quality degradation (Van Meter et al., 2018; Hansen et al., 2021). In regions like the Midwestern United States and Northwestern Europe, agricultural intensification often involves considerable land improvement with a distinctive feature: artificial tile drainage systems (Gramlich et al., 2018; Hejase et al., 2022). While extensive artificial tile drainage systems benefit crop growth (Mourtzinis et al., 2021), they also accelerate the transfer of nutrients from the land into the receiving water bodies, leading to increased incidence of eutrophication and harmful algal blooms (HABs) (Castellano et al., 2019; Ren et al., 2022). Since the early 2000s, phosphorus (P) loss through tile-drained agricultural fields has been recognized as an issue for large watersheds due to the wide application of subsurface drainage systems and no-till farming (King et al., 2015; Jarvie et al., 2017; Singh et al., 2023). In Western Lake Erie (USA), for example, subsurface tile drainage networks account for 49 % of the

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soluble P loss and 48 % of the total P (TP) exports from the watershed (Smith et al., 2015). While there is increasing evidence that implementation of conservation practices can reduce the TP and particulate P, they are less effective in removing dissolved reactive phosphorus (DRP), a significant factor contributing to HABs, is known to be less pronounced (Scott et al., 2023).

P removal structures installed at the edge of fields are an effective technology for removing DRP from drainage/runoff, thereby mitigating the impact of P loss from intensified agricultural activities (Penn et al., 2014; Mendes et al., 2022; Scott et al., 2023). These structures use a variety of phosphorus sorption materials (PSMs), including natural minerals, by-products (e.g., metal shavings, fly ash, and steel slag), and synthetic materials (Zhou et al., 2022; Ai et al., 2023). Although some PSMs, such as metal shavings and steel slag, have been shown effective performance in field applications (Penn and Bowen, 2017), the cost, maintenance, and disposal of spent sorbents hinder their wide adoption. So far, research has yet to reach a consensus on utilizing industrial wastes, such as slags, as PSMs in agricultural fields (Wang et al., 2021). Therefore, addressing these challenges and improving the implementation of PSMs are of paramount important for DRP loss management.

Biochar has demonstrated a potential for capturing DRP from contaminated water (Zhang et al., 2020; Yang et al., 2021). Our previous studies designed a lime sludge-based biochar and a gypsum-incorporated biochar, achieving DRP removals of up to 76 % and 100 % in laboratory batch experiments, respectively (Yang et al., 2021; Katuwal et al., 2023). Similarly, Mehrabinia et al. (2022) observed that active nano-biochar can remove over 47.8 % of P from agricultural drainage. Biochar could also improve soil health and crop production (Bai et al., 2022; Ye et al., 2020). Thus, biochar is a more environmentally safe and friendly material compared to many PSMs, especially industrial wastes. Land application of spent biochar into nearby farms can not only retain nutrients within a closed agricultural loop, but also reduce disposal costs (Zhou et al., 2023). Therefore, biochar could be an emerging alternative to traditional PSMs, renovating *in-situ* nutrient loss reduction technology. Despite its potential, real-world field applications of biochar are scarcely studied. Most studies relied on short-term laboratory batch sorption or columns to assess biochar capacities DRP capture, which may not fully understand biochar's effectiveness in practical settings (Zhang et al., 2020; Zhou et al., 2024). To date, there has been no comprehensive analysis including removal performance, field feasibility, economic consideration, and environmental impacts to explore the engineering-scale biochar application on the *in-situ* DRP removal.

In this study, we developed designer biochar pellets tailored for real-field application. The objectives of this study aimed to (*i*) evaluate the effectiveness of designer biochar pellets in reducing DRP from tile drainage water under a field-scale demonstration; (*ii*) identify the key factors influencing the DRP removal of designer biochar pellets; (*iii*) assess the application potential and environmental benefits of this work using techno-economic analysis (TEA) and life cycle assessment (LCA); and (*iv*) outline a holistic sustainable management paradigm to boost the adoption of biochar pellet, benefiting stakeholders, environmental agencies, and farmers. The findings of this study provide first-hand information on the engineering application of biochar for water quality improvement, offering an in-depth understanding of the costs and benefits of this practice for DRP loss management. In addition, this study is expected to serve as a foundational reference for integrating biochar into a variety of drainage systems, such as bioretention basins, urban drainage, bioswales, and constructed wetlands (Chen et al., 2021).

2. Materials and methods

2.1. Manufacturing designer biochar pellets for engineering application

In this study, pelletized designer biochar were manufactured from powdered biochars that have previously demonstrated effective DRP removal in laboratory experiments (Yang et al., 2021). Two types of designer biochar pellets with particle sizes ranging from <1 cm to 2∼3 cm (Fig. S1) were produced under a pelleting mill (MILL-10 Pellet Mill 10HP, Colorado Mill Equipment, USA). Pelleting technology is commonly used to facilitate the management of different agricultural inputs that are otherwise in an amorphous powder form (Lollato et al., 2013). According to preliminary experiments, two types of pelletized designer biochar with particle sizes ranging from <1 cm to 2∼3 cm were selected and manufactured at scale for field experiments (Fig. S1). The main ingredients of the designer biochar pellets include biochar, lime sludge, and binding agents (i,e., clay). The dry mass elemental components of the designer biochar pellet were 22.50 % C, 0.74 % H, 0.07% N, 9.59 % O, and 70.53 % ash, respectively. The Brunauer-Emmett-Teller (BET) surface area of the designer biochar was 150.78 m^2/g (determined by nitrogen adsorption).

The experiments were conducted at the Metropolitan Water Reclamation District of Greater Chicago (MWRDGC) Fulton County site (40 ̊28′38.99″N, 90 ̊6′10.75″W), IL, USA. Details on the field condition and agricultural activities are shown in Supplementary Material Text 1. Briefly, field demonstration was conducted in a 9.71 ha (24 acres) field at the research site, and divided into two equal subfields (i.e., Field A and Field B). In each subfield, drainage tiles were installed 1 m below the soil surface at intervals ranging from 8 to 40 m with existing woodchip bioreactors well-constructed in the main drainage outlets to remove nitrate from drainage water. In each subfield, a biochar-sorption chamber (P removal structure, Length: 35 cm; Width: 25 cm; Height: 25 cm) (Fig. S1) was designed and constructed in the main tile drain to allow drainage water to pass through (Fig. S2). Woodchip bioreactors are widely implemented for nitrate reduction in agricultural drainage systems and have been installed at the studied fields. To examine the effect of woodchip bioreactors on DRP capture by biochar, in Field A, the biochar-sorption chamber was installed before the woodchip bioreactors, whereas, in Field B, the biochar-sorption chamber was installed downstream of the woodchip bioreactors. The experiments were operated in two phases: Phase 1 (May 13, 2021 - November 4, 2021, total 170 days) and Phase 2 (November 4, 2021 - July 12, 2022, total 250 days) (Table S1). In Phase I, approximately 10 kg of designer biochar pellets with uniform particle sizes of 2-3 cm were loaded into the biochar-sorption chambers and then placed into the tile drainage system (Fig. S1). In Phase II, the spent designer biochar pellets were replaced by about 13 kg of small-size designer biochar pellets (particle sizes <1 cm).

2.2. Field monitoring and analysis

To systematically evaluate the performance of designer biochar pellets in reducing DRP loss, the following indices were explored: (i) cumulative DRP load (kg), (ii) the DRP capture amount (mg DRP/g biochar/day), and (iii) DRP removal efficiency (%). Cumulative DRP load reduction is calculated by summing up the daily differences between the inlet and outlet DRP loads over the entire phase. DRP removal efficiency is calculated by the ratio of the difference between the inlet and outlet DRP concentrations to the inlet DRP concentration, expressed as a percentage. To determine the biochar DRP removal efficiency, DRP concentrations were analyzed from water samples collected before and after biochar-sorption chambers (Details in Supplementary Material Text 2).

Typically, a more dynamic pattern occurs in the field compared to the laboratory experiments, in which multiple factors can influence the performance of designer biochar pellets. In this study, these influences are categorized into two main factors: human factors and environmental factors. Human factors primarily involve elements intentionally influenced or altered by engineers that may potentially impact biochar pellet performance, including biochar pellet sizes as well as the relative position of biochar-sorption chamber and woodchip bioreactors. Environmental factors include drainage flow rate, influent concentrations, and climate parameters such as relative humidity, temperature, and precipitation. Detailed methods to obtain daily and overall nutrient loads can be found in our previous report (Oladeji et al., 2023).

2.3. Economic and environmental assessment

Techno-Economic Analysis (TEA) and Life-Cycle Assessment (LCA) were performed to evaluate the economic viability and environmental benefits of the proposed technique, which is crucial for promoting large-scale applications. For TEA, the cost analysis was divided into two main categories: (a) production costs of designer biochar pellets (e.g., capital, operation, and materials), and (b) the DRP removal cost using biochar-sorption chamber at the field scale (e.g., structure construction cost, materials cost, and installation, operation, and maintenance costs). The benchmark case considered a 40 % cumulative DRP removal over a 15-year horizon at the 10 % discount rate for removal structures. Moreover, stochastic simulations were performed to estimate the impact of eight uncertainty variables. Further, analyses of DRP removal and cost efficiency under various field strategies and biochar replacement scenarios based on benchmark assumptions were conducted to inform realistic future implementation strategies.

The LCA aimed to: (i) evaluate the environmental performance of using designer biochar pellets for DRP removal, focusing on energy consumption expressed in megajoules (MJ) and Global Warming Potential (GWP, kg CO_2 eq) throughout the entire process from the production of designer biochar pellets to their application in tile-drained fields for DRP removal, and reuse of spent biochar pellets as soil amendments; and (ii) adopt the technology strategically, by conducting scenario analysis and stochastic simulation aligning with TEA results. The scope and functional units of this study focused on the cradle-to-grave environmental performance of removing 1 kg of DRP using designer biochar pellets. Detailed descriptions of the calculation processes and assumptions of TEA and LCA can be found in Supplementary Material (Text 3 and Text 4).

2.4. Reuse of spent designer biochar pellets as soil amendment

To explore the feasibility of spent designer biochar pellets as a soil amendment, radish (*Raphanus sativus* var. 'Cherry Belle') cultivation experiments were conducted (Supplementary Material Text 5).

2.5. Statistical analysis

The Spearman's correlation method was employed to analyze the relationships between variables. The correlation between seven input parameters in the DRP removal efficiency and DRP removal rate of the system has been analyzed. The description of dataset for correlation analysis is provided in Supplementary Material Text 2. Significant differences were identified based on a $p < 0.05$ using R v 4.3. Additionally, figures were generated with Origin 9.0 software (Origin Lab Corporation, USA).

3. Results and discussion

3.1. Site hydrology and DRP losses from fields

The precipitation and drainage flow at the research sites during the experimental periods (Phase I and Phase II) are shown in Fig. S3. Both tile-drained experimental fields (Field A and Field B) experienced typical seasonality with higher flow before the growing season and low to no-flow during the growing season. Drainage rates measured during the study periods varied between 0 and 0.3 cm/day at the two sites (Fig. S3). During both experimental periods, Field A had a higher overall drainage flow rate compared to Field B. Throughout Phase I, the total drainage flows from Field A and Field B were 100 mm and 50 mm which accounted for 13 % and 6.52 % of the total precipitation, respectively. These results suggest that the majority of rainwater was either lost via evapotranspiration or used by plants during the growing season. In contrast, the total drainage flows during the Phase II experimental period were 372.1 mm and 259.5 mm for Field A and Field B, respectively. Unlike Phase I, Phase II included a non-growing season as well as a growing season. Most precipitation loss via drainage occurred before the growing season, especially in the early spring due to snowmelt and storm runoff.

Fig. 1 shows the daily and cumulative DRP losses from two experimental field sites. Concentrations of DRP in the drainage water during the experimental periods ranged from 0.01 mg/L to 0.55 mg/L for Field A and 0.01 mg/L to 0.41 mg/L for Field B, with flow-weighted mean concentration of 0.13 mg/L and 0.08 mg/L for Field A and Field B, respectively. During Phase I, cumulative DRP loss of Field A (1.56 kg) was much higher than Field B (0.34 kg). Similarly, in Phase II, DRP loss was higher for Field A (1.53 kg) compared to Field B (0.96 kg). The cumulative DRP losses were consistent with the difference in drainage flow at the two sites, as higher drainage flow rates and DRP losses were observed in Field A than in Field B in both Phase I and Phase II periods.

3.2. Field performance evaluation of designer biochar pellets in Phase I and Phase II

Results of cumulative DRP load reduction, DRP removal efficiency, and biochar DRP capture amount are shown in Fig. 2 for both Phase I and Phase II experiments conducted in Field A and Field B. During Phase I, the cumulative DRP losses from Field A and Field B after biochar chamber treatment were around 1.51 kg and 0.31 kg (Fig. 2a), resulting in a reduction of 0.05 kg and 0.03 kg in DRP loads from tile drainages in Field A and Field B, respectively. Accordingly, the capture amounts by the biochar pellets were 5.2 mg g^{-1} (DRP/biochar) and 3.0 mg (DRP/biochar) for Field A and Field B, respectively (Fig. 2b). As shown in Fig. 2c, the median DRP removal efficiencies by designer biochar pellets were higher in Field A (12.0 %) compared to Field B (1.3 %). These results indicates that the designer biochar pellets with the particle size > 2 cm had a low efficiency to remove DRP from drainage water in Phase I.

At the beginning of Phase II, the spent biochar pellets (particle size 2-3 cm) were replaced with new designer biochar pellets (particle sizes < 1 cm). Fig. 2 shows a higher performance in DRP reduction for new designer biochar pellets in Phase II compared to the biochar pellets used in Phase I. In Field A, the cumulative DRP load decreased from 1.53 kg to 1.13 kg, while in Field B, a reduction of cumulative DRP load from 0.96 kg to 0.61 kg was observed (Fig. 2a). The smaller particle size of designer biochar pellets achieved DRP capture amounts of 30.4 mg g^{-1} (DRP/biochar) and 27.2 mg g^{-1} (DRP/biochar) for Field A and Field B, respectively (Fig. 2b). Meanwhile, the DRP removal efficiency of the designer biochar pellets exhibited a substantial increase (Fig. 2c), reaching up to 38 % in Field A and 41 % in Field B.

The low DRP removal efficiency of designer biochar pellets in Phase I might be related to the short retention time of drainage water flowing through the biochar-sorption chambers, resulting in insufficient time for interaction between DRP and designer biochar pellets. The improved performance in Phase II may be attributed to the biochar pellets with smaller particle sizes (<1 cm), indicating biochar particle size plays a significant role in contaminant removal (El Hanandeh et al., 2017; Bian et al., 2024). In this study, both biochar pellets were manufactured from lime sludge-incorporated designer biochar. Our previous study revealed that the main adsorption mechanism of DRP by the lime sludge-incorporated designer biochar is chemical precipitation (Yang, et al., 2021). When DRP species (i.e., $H_2PO_4^-$ and HPO_4^{2-}) in nutrient-containing water reach the alkaline surface of the designer biochar, they can readily convert to $H_2PO_4^-$ and HPO_4^{2-} and PO_4^{3-}. These two DRP species could readily react with metal ions (i.e., Ca^{2+} and Mg^{2+}) incorporated on the designer biochar surfaces to generate $Ca_3(PO_4)_2$, $CaHPO_4$, and $MgHPO_4$ precipitates (Yang, et al., 2021; Katuwal et a., 2023). Compared to the large particle size, small particle sizes facilitate DRP species access to the interior of biochar pellets,

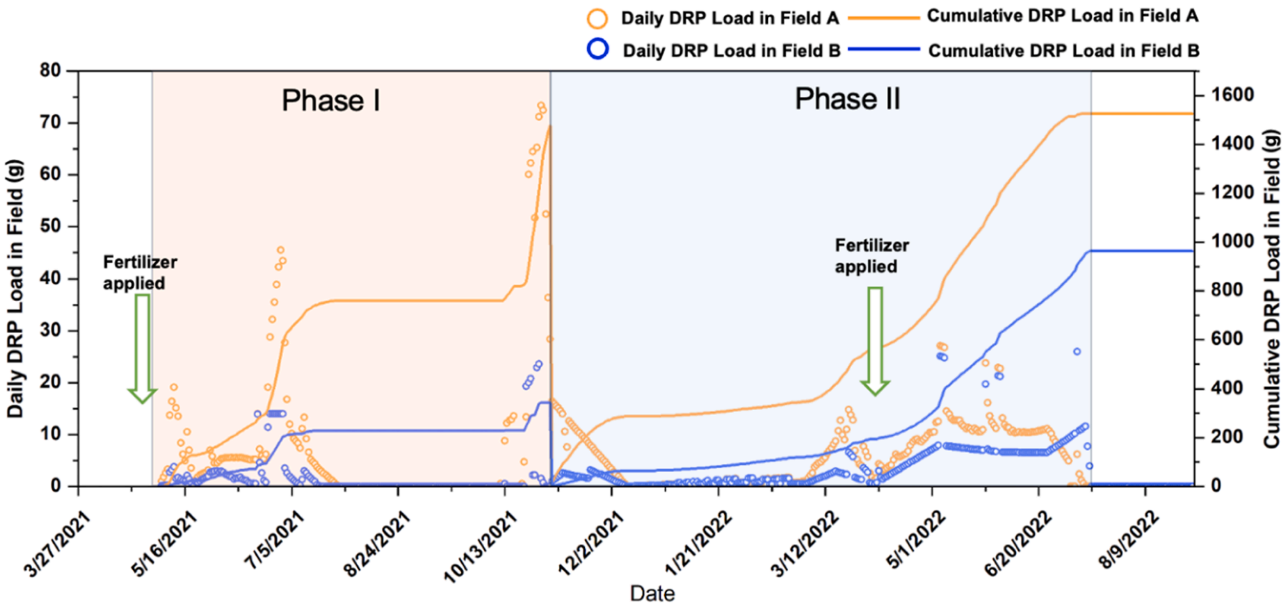

Fig. 1. Daily and cumulative DRP load measured in the main drain lost from tile-drain agroecosystems before biochar chamber treatment.

resulting in faster precipitation reactions between DRP and metal ions incorporated on designer biochar. Thus, the small particle size of biochar pellets means faster sorption kinetics for DRP. To further support this hypothesis, the effect of biochar pellet particle sizes on the removal of DRP was investigated using a laboratory sorption experiment (Fig. S5). The lab results demonstrate that designer biochar pellets with small particle sizes spend less time reaching the sorption equilibrium (Fig. S5). For example, the designer biochar with a 2.0 cm pellet size needed 4 h to reach its sorption equilibrium, while the biochar powder with <0.2 cm particle size just needed 10 s for DRP precipitation on designer biochar. According to our field observations, the residence times in the biochar-sorption chamber are <30 s in Phase I and 60 s in Phase II, respectively. The smaller biochar pellets used in Phase II result in a relatively longer residence time for DRP capture by designer biochar compared to Phase I.

From a DRP removal perspective, designer biochar powders exhibit better performance in DRP loss reduction since it requires less time to capture DRP from drainage water. In real-field applications, however, biochar powders or pellets with very small particle sizes would encounter some issues, especially in flow-through drainage systems. The powdered designer biochar could be readily washed away from biochar-sorption chambers by drainage flow. If biochar with very small particle sizes is used, the biochar-production chambers could be readily clogged since the drainage water may carry sediments. Using smaller-size biochar pellets in Phase II showed that a deliberate trade-off between structural design and removal performance must be achieved to optimize the application of biochar to mitigate DRP loss in tile drainage systems.

In addition, total P removal was also monitored in both fields for each phase (Fig. S3). As shown in Fig S4a, the cumulative total P load in Phase I decreased from 3.89 kg in influent to 2.63 kg in effluent for Field A and from 1.92 kg to 1.57 kg in Field B. During Phase II, total P was reduced from 4.49 kg to 2.37 kg in Field A and 2.75 kg to 1.15 kg in Field B (Fig. S4b). Compared to DRP removal (Fig. 2a), the reductions of total P are higher in both phases and fields, suggesting that the designer biochar pellets not only adsorb dissolved P, but also capture suspended P.

3.3. Muti-Factors affecting the DRP loss reduction using designer biochar pellets

Changing climate may increase the incidence of high-intensity rainfall events which could affect flow-through dynamics and temperature in the drainage channels. Fig. 3 represents the correlation between individual human and environmental factors as well as their impacts on the biochar DRP reduction performance during the field demonstration, evaluated by the Spearman correlation coefficient (ρ).

The DRP load reduction rate showed a significantly positive relationship with influent DRP concentration (ρ = 0.51) and a mildly positive correlation with temperature (ρ = 0.26) (Fig. 3). This result indicated that more DRP amounts could be captured by the designer biochar at higher temperatures as well as higher influent DRP concentrations. On the contrary, the average DRP removal efficiency showed an inverse relationship with influent DRP concentration ($\rho = -0.50$) and temperature ($\rho = -0.61$). DRP removal efficiency by the designer biochar was notably higher (median = 50.9 %) when the influent DRP concentration ranged from 0 to 0.1 mg/L. As the influent concentration increased, removal efficiency gradually decreased, reaching median efficiency of 28.28 % at 0.1–0.2 mg P/L, 25.71 % at 0.2–0.3 mg P/L and 5.71 % at greater than 0.3 mg P/L (Fig. S6). These field-scale observations corroborate laboratory results, in which the DRP removal rates by biochar decreased with increasing initial DRP concentrations (Feng et al., 2021; Biswas et al., 2023). The negative relationship between temperature and average DRP removal efficiency may be attributed to high correlation ($\rho = 0.75$) of the field temperature with the influent DRP concentration (Fig. 3). Field data demonstrated that a lower flow rate, or even no flow, often occurs in the drainage system during the growing season when temperatures are relatively higher. During this period, an elevated concentration of DRP in the drainage system was observed, especially after the fertilization application period (Fig. 1).

The DRP load reduction rate showed slight correlation with the drainage flux ($\rho = 0.19$) and precipitation ($\rho = 0.09$) while the role of relative humidity was not significant on the removal indicators. Although the correlation between daily precipitation and the removal indicators was not strong, studies suggest that rainfall events could become a driver to impact drainage flow dynamics and inflow DRP concentrations (Vidon and Cuadra, 2011; Madison et al., 2014). This warrants further event-based analysis in the future to better understand the climate change impacts.

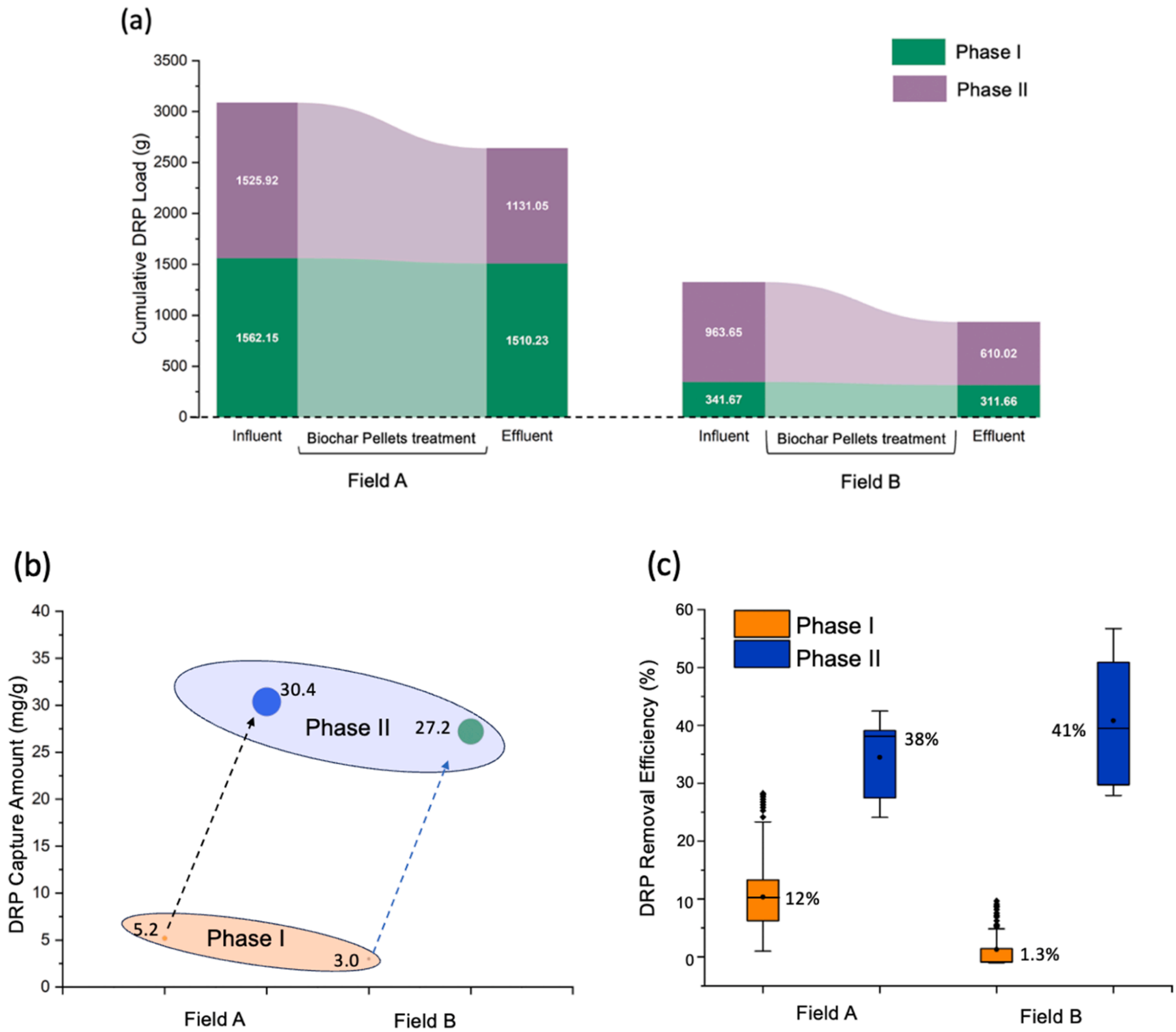

Fig. 2. Comparison of designer biochar pellet DRP removal performance under two particle sizes in two research sites during Phase I and Phase II periods: (a) The cumulative DRP load reductions using the designer biochar pellets in Phase I and Phase II; (b) DRP capture capacities by different particle size biochars in Phase I and II; (c) The daily average DRP removal efficiency at different biochar particle pellets in Phase I and Phase II.

Notably, out of the human factors, the DRP load reduction rate was correlated to biochar particle size ($\rho = -0.17$) and showed a low degree of correlation with the biochar position ($\rho = 0.08$). This implies that the use of smaller pellet sizes could be crucial to achieve higher DRP removal and the addition of biochar sorption chambers after the bioreactors could be advantageous. The enhanced DRP removal using smaller pellet sizes biochar has been well-documented as we discussed in Section 3.3. The addition of biochar sorption chambers after bioreactors potentially benefits DRP removal as the bioreactors can act as a buffer to provide more stable drainage flow rates throughout the biochar systems. This stability is essential for optimizing the contact between the drainage water and biochar pellets, ensuring consistent treatment. However, these values should be considered with more uncertainty in the field, as represented by the higher ranges of confidence interval (Fig. 3). These analyses were also further supported by the *t*-tests which demonstrated significantly higher ($p < 0.05$) mean removal efficiency for biochar pellet sizes and biochar sorption chamber positions (Table S2 - S3).

Significant research has highlighted the importance of flow rate, inlet DRP concentration, and filter material retention mechanisms for long-term filter efficiency (Pugliese et al., 2023). In this study, DRP removal by biochar was impacted by environmental factors (e.g., influent concentration, drainage flux and precipitation) and human factors (e.g., size of biochar pellets, and biochar structure position) to some degree. Under field conditions, it is more challenging to control inlet P concentration and drainage flux, thus strategic engineering applications are necessary to achieve higher DRP removal.

3.4. Techno-economic analysis of designer biochar pellets for DRP loss reduction

The techno-economic analysis was performed to explicate the economic feasibility from the manufacturing designer biochar pellets to their field application to remove DRP from drainage water. As shown in Table S4 - S6, the average production cost of designer biochar pellets was \$412.64/ton which was close to the lower range of commercially available biochar costs ranging between \$449 to \$1,847 (Nematian et al., 2021) and much less than the market cost of granular activated carbon (GAC) (\$800- \$ 2,500/ton) (He et al., 2022). The cost advantage of our designer biochar pellet production was due to the inexpensive raw

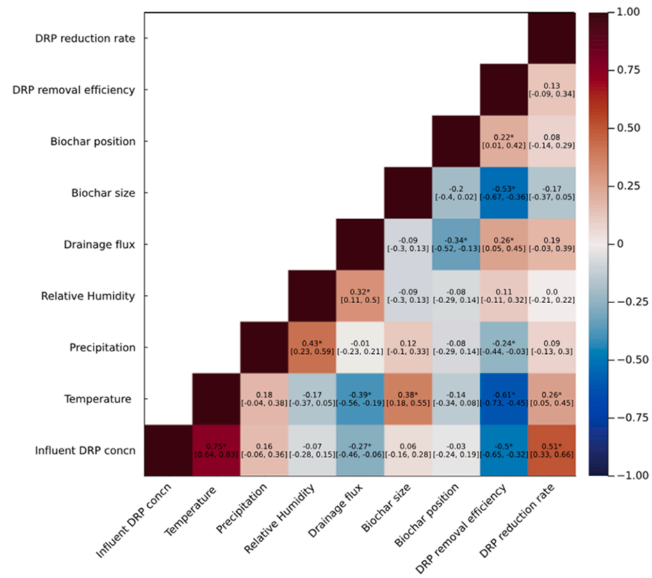

Fig. 3. Spearman correlation coefficient of system output parameters: (efficiency and reduction rate of DRP) with system input parameters: influent DRP concentration, Temperature, Precipitation, Relative Humidity, drainage flux, Biochar position, and Biochar size. *Represents statistically significant data at level ($p < 0.05$) and the numbers inside brackets represent the uncertainty in correlation (values at 95 % confidence interval).

materials and no additional activation procedures compared to GAC production. The operation, materials, and capital costs contributed 71.81 %, 19.34 %, and 8.85 %, respectively, to the pilot-scale production of the designer biochar pellets.

For benchmarking TEA of the designer biochar-based DRP removal at the field-scale application, we adopted the 15-ha field with a total DRP input of 8 kg/year (~ 0.5 kg/ha/year) into the structure which could practically remove 40 % DRP with the replacement of biochar every year over the design life of the structure of 15 years. Under the benchmark assumptions, the structure can remove a cumulative 48 kg DRP over 15 years (3.2 kg DRP removal each year) at the Present Value (PV) cost of $7,932 with an average unit DRP removal cost at PV of $165/kg DRP (Table S7) at the discount rate of 10 % throughout the study horizon. However, the estimated annual average cost incurred each year of the design period was $326/kg DRP. It should be noted that the annualized capital cost was evenly distributed over the study horizon at the 10 % discount rate (benchmark rate) to demonstrate the annual average unit cost over the years. The variation in estimated removal cost over the years was demonstrated at different inflation rates (Fig S7a) and the cost of biochar (Fig S7b). Considering the inflation over the years (Fig S7a), the maximum annual average removal cost could reach as high as $1,215/kg DRP at the 20 % inflation rate, while considering the variability of biochar cost, average removal cost ranged from $301/kg DRP to $329/kg DRP (Fig S7b).

The costs of different designer pellet replacement scenarios were further envisioned to guide practical application, considering the saturation of biochar pellet. It was assumed that the removal efficiency of DRP would be consecutively reduced by 5 % each year (for example, 40 % for the first year, 35 % for the second year, 30 % for the third year, and so on) until replacement. Under the different replacement scenarios (Fig S8b), the actual annual cost of DRP removal averaged $308 to $352/kg DRP with PV ranging from $6,510 (replacement every five years) to $7,931(annual replacement). These results demonstrated that, while opting for biochar replacement every five years resulted in lower associated costs at PV, cumulative DRP reduction decreased from 48 kg to 36 kg in this scenario (Fig S8c). Therefore, the tradeoffs indicate that the most cost-effective scenario would be replacement every two years with an average cost of $308/kg DRP removal, closely followed by $314/kg DRP for replacement every three years. Under the biennial replacement scenario, the system removed 45 kg of cumulative DRP over 15 years as compared to 48 kg with replacement every year and 42 kg with replacement every three years (Fig. S8c-S8d).

Moreover, the stochastic simulation under 2000 different combinations of the parameters showed that the total cost of the DRP removal structure at PV ranged from $6318 to $8941 with an average of $7304, wherein the average capital cost of the structure, cost of materials, operation, and maintenance comprised 77.55 %, 14.81 % and 7.64 % of the total cost (Table S8). Similarly, the mean of annual DRP removal cost was $359/kg DRP ($160 /kg DRP at PV) and ranged from $68 /kg P to $899/kg DRP (Fig. 4a). Under the stochastic simulation of different replacement scenarios, the average unit cost of DRP removal was least ($348/kg DRP) for replacement every three years and highest for replacement every year ($403/kg P) (Fig. 4b). The average unit cost of removal for replacement every year was significantly higher ($p < 0.05$) compared to other scenarios, however, the average cost of removal for replacement every three years was not significantly lower at ($p < 0.05$) compared to replacement every two, four or five years. Thus, biochar replacement every two years is recommended as the optimum option, considering the tradeoff between DRP removal and the cost of removal. It is because the replacement of biochar every two years also offered higher DRP removal compared to that of every three years and cost significantly less than replacement every year. This conclusion also reinforced our finding at the benchmark assumption (Fig. S8d). The

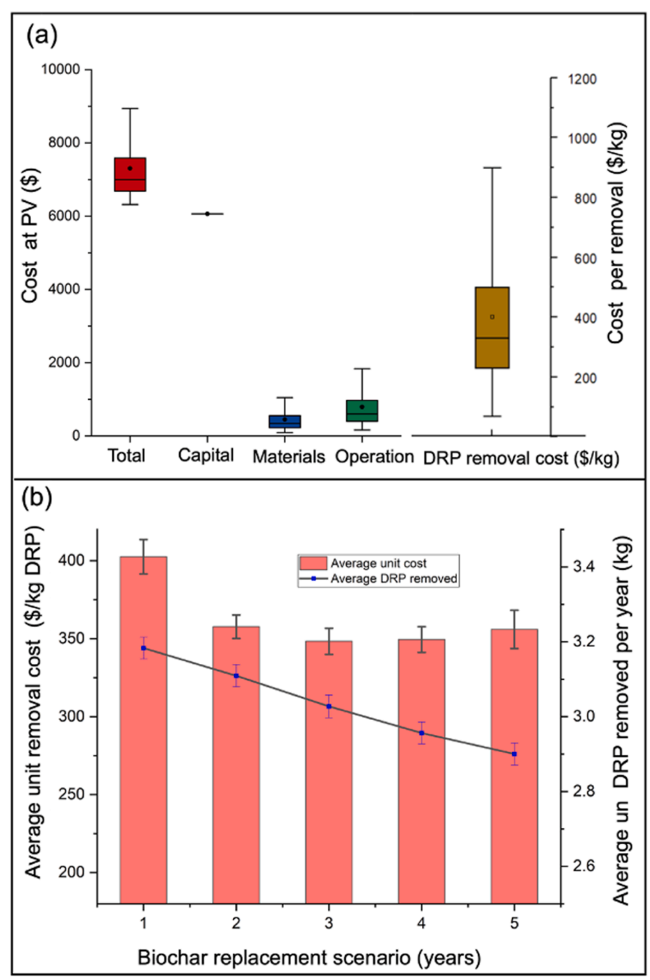

Fig. 4. a) Breakdown of DRP removal cost under stochastic simulations b) Annual average removal and average unit removal cost under stochastic simulations.

above-estimated costs are comparable to the existing DRP removal techniques which range from $ 100 - $1,300/kg DRP removal (Scott et al., 2023).

3.5. Life- cycle assessment of designer biochar pellets for DRP loss reduction

Fig. 5 shows the total net positive energy (1300 MJ) and total net negative Global Warming Potential (GWP, -170 kg CO_2 eq) of reducing 1 kg of DRP loss from the tile-drained field using designer biochar pellets. Biochar energy, including the energy consumed for pelleting and pyrolysis using electricity and energy produced during pyrolysis (heat products and stable carbon), was the major contributor to energy net production (2000 MJ/kg DRP removal) and GWP net reduction (-200 kg CO_2 eq/kg DRP removal). Moreover, the disposal of spent biochar pellets for the soil amendment, represented by avoided nitrogen, phosphorus, and potassium fertilizer productions and emissions, also contributed to production of energy and the reduction of GWP (-2.6 kg CO_2 eq/kg). The total GWP of 1 ton of biochar pellets produced in this study, from cradle to grave, was -2.6 tons CO_2 eq, with negative values representing the greenhouse gas emissions abatement. This result was similar to the findings from previous studies (-12.5 to -2.6 tons CO_2 eq) (Roberts et al., 2010; Gaunt and Cowie, 2012), but closer to the higher end of the range mainly due to two factors. First, the feedstocks used in their study were agricultural products, such as wheat stover or yard waste, which have lower GWP than the waste materials used in this study. Second, the energy used for pyrolysis in this study was fossil fuels powered electricity and not fossil fuels, making the energy consumption and emissions for the pyrolysis process higher than previous studies (Roberts et al., 2010).

In alignment with TEA, we evaluated the environmental impacts of different biochar pellets replacement scenarios. As shown in Fig. S9, it is not surprising that replacing the biochar annually achieved the highest annual emissions abatement (-170 kg CO_2 eq) as well as the highest cumulative DRP removal (Fig. S8a); however, it was also the most expensive scenario (Fig. S8c). The GWP of the two-year replacement scenario was almost half of the one-year replacement scenario because less biochar is needed in the system every two-year period. The trend in DRP removal efficiencies of the replacement scenarios with more than one year also appeared to be a wave instead of a straight horizontal line like the one-year replacement scenario. This is because the DRP removal efficiency decreased as the biochar remained in the P removal structure for more than one year. The GWP overlaps occurred among replacement scenarios of three to five years. On the one hand, greenhouse gases reduction of biochar production divided by years makes the greenhouse gases reduction of each year in the five-year scenario less than that in the three-year scenario. On the other hand, the DRP removal efficiency was 5 % lower than the previous year, which made the GWP of the first year of the three-year scenario higher than the GWP of the last year of the fourth year and the GWP of the last year of the fifth-year scenario. Therefore, practitioners and farmers should consider the results synergistically with the results from TEA scenario analysis - the lowest cost for P removal occurred at replacement once every three years. Of these options, replacing biochar once every two years could be the careful middle road that maximizes the sum of the two outcomes. The GWP resulting from the stochastic simulation (Fig. S10) further demonstrated the emissions abatement potential of using designer biochar pellets for DRP reduction, although the uncertainty of the abatement was high (ranging from -12 to -200 kg CO_2 eq per kg DRP removal), in which the major uncertainty of GWP over its lifetime came from the GWP of materials (biochar production).

3.6. Environmental implication

3.6.1. Potentials to boost technology adoption

If designer biochar pellets are employed in practice, we must consider how to bring benefits to users and stakeholders to boost technology adoption. In Section 3.4, we demonstrated that the application of designer biochar pellets for DRP loss reduction is economically viable. We further investigated the feasibility of reusing spent designer biochar pellets as a soil amendment for crop production. Previous studies have fully demonstrated the benefits of biochar application for soil health and crop growth (Bai et al., 2022; Ye et al., 2020). A complementary experiment has been conducted to examine the reuse of spent designer as a soil amendment on radish production. The results (Fig. S11) not only show higher radish weights after using 2 % of spent designer biochar compared with the control group (increased by 15.57 % to 30.76 % by weight) but also suggest the possibility of reducing fertilizer application while maintaining comparable or better agronomic yields. Farmers can reduce their fertilizer costs if spent biochar pellets benefit their crop production. The potential for cost savings is a significant motivator for farmers to embrace this technology. The co-location of expanded tile drainages and surrounding drained croplands offers an immediate opportunity to use biochar pellets and reuse the spent biochar pellets, instead of landfilling, which can further solve disposal costs and subsequent management issues of spent materials. Biochar pellets are easy to replace and apply to the soil without dust emissions. Most biochar has a lower bulk density (<0.6 g cm^{-3}) than soil (~1.25 g cm^{-3}),

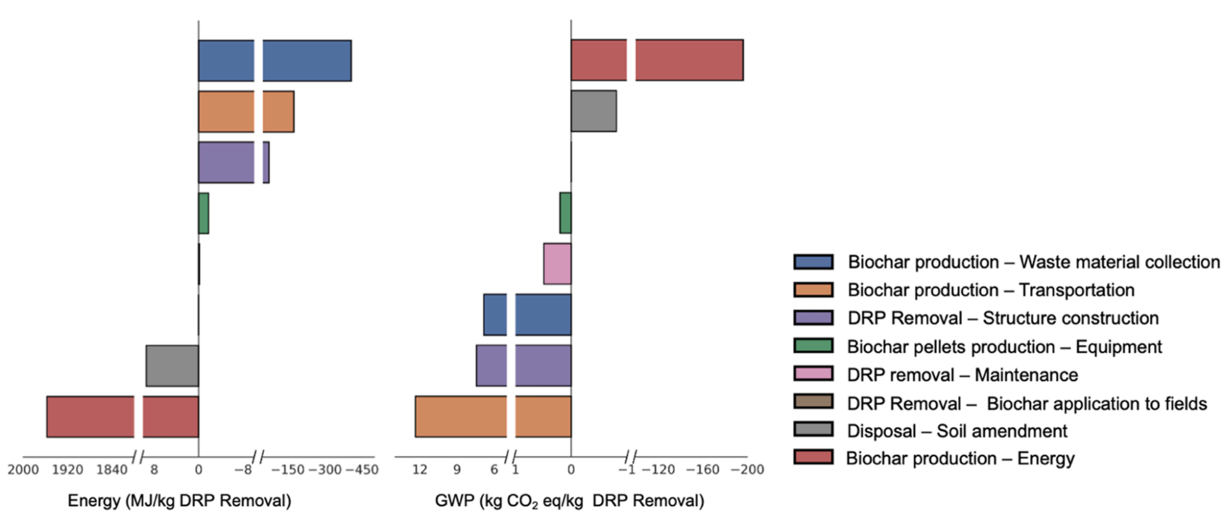

Fig. 5. Contribution analysis for the different steps to net energy and climate change impact of removing 1 kg of DRP from tile-drainage systems using designer biochar pellets at field scale.

which is to float on the soil surface and be easily carried away by runoff or blown away by wind, while designer biochar pellets with a higher bulk density can be integrated into the soil. Pelletized soil amendments can potentially be applied in furrow at planting together with the seed (Lollato et al., 2013) as starter fertilizers, although the feasibility of this practice is yet to be evaluated and could be a focus of future studies. Therefore, a systematic pathway that integrates waste management, biochar pellets for nutrient loss reduction, and reusing spent biochar pellets can be envisioned to mitigate nutrient pollution and improve water quality.

3.6.2. The way forward

The optimization of system design to further improve the effectiveness of nutrient loss reduction is another key area where we recommend further research. In this study, a longer contact time is needed to allow more effective interaction between the biochar pellets and DRP in drainage water. However, it is important to recognize that under field conditions, various competing chemical and biological processes may inhibit the effectiveness of biochar for DRP removal. For instance, competing ions could significantly reduce the adsorption capacity of biochar, leading to lower removal efficiencies than those observed in controlled laboratory experiments. Engineers can design more advanced structures to enhance the DRP removal efficacy by accounting for these inhibitory factors. For instance, one promising avenue is the implementation of multiple stages of biochar sorption channels or baffle structures to promote longer contact times between biochar and nutrient-rich water. These designs could also incorporate mechanisms to mitigate the impact of competing processes, such as pre-treatment steps to reduce the presence of competing ions or the integration of microbial management strategies to maintain optimal biochar performance. More investigations are required to test these applications in long-term studies (longer than one year in real situations) to increase efficacy, particularly under diverse environmental conditions that reflect the complexities of field scenarios. Practically, such structures are suited well into existing filter strips located along ditches and streams and combined with other edge-of-field technologies. Additionally, considering the above-mentioned inhibition factors, expanding research to include field trials that assess the real-world performance of these optimized designs, will be crucial for improving nutrient loss reduction at a practical scale (Zhou et al., 2024).

4. Conclusion

The utilization of designer biochar pellets offers water quality managers and agricultural practitioners a promising avenue to reduce DRP from tile-drained agroecosystems and promote sustainability. This study should help biochar research move beyond proof-of-concept to include the perspectives of engineering application, economic and environmental consideration. Specific conclusions of this study are:

1. Compared with designer biochar pellets with particle sizes of 2-3 cm in Phase I, the results showed a comprehensive DRP loss reduction improvement in terms of cumulative DRP load reduction, DRP removal efficiency, and the DRP capture amount in Phase II using designer smaller-sized biochar pellets (particle sizes of < 1 cm). In the field application, biochar pellet performance is impacted by multi-factors including pellet size, position of structure, influent DRP concentrations, and drainage flow.
2. The designer biochar pellets were found to be economic with an average production cost of $413/ton. Under the benchmark assumptions, the average unit cost of removal was $326/kg DRP. Moreover, under wide assumptions of economic parameters such as inflation, discount rates, cost of biochar, and project life; and system design parameters such as Phosphorus input, removal efficiency, and replacement scenarios, the unit cost of removal was $\$359\pm177$/ kg DRP. To guide practical application, the different replacement scenario analysis indicated that the most cost-effective scenario for maximizing DRP removal was biochar pellet replacement every two years with an average cost of $358/kg DRP.
3. Life cycle assessment suggested that the entire process can achieve benefits beyond nutrient loss reduction, including carbon sequestration (CO_2 emissions abatement ranging from -12 to -200 kg CO_2 eq/kg DRP removal), soil amendment, energy production, and reduction of eutrophication potentials.
4. Future research will focus on optimizing strategies and scalability for biochar and DRP removal structures to accommodate sustainable urban and agricultural drainage systems and support policymaking in water quality management.

CRediT authorship contribution statement

Hongxu Zhou: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Haribansha Timalsina:** Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Conceptualization. **Peng Chen:** Writing – review & editing, Writing – original draft, Investigation, Formal analysis. **Sophie Circenis:** Writing – review & editing, Investigation. **Richard Cooke:** Writing – review & editing, Resources, Investigation. **Olawale Oladeji:** Writing – review & editing, Investigation. **Guanglong Tian:** Writing – review & editing, Investigation. **Romulo P. Lollato:** Writing – review & editing, Investigation. **Rabin Bhattarai:** Writing – review & editing, Investigation. **Wei Zheng:** Writing – review & editing, Writing – original draft, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2024.122500.

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