



Meta-analysis and empirical research on the effectiveness of biochar in remediating tetracyclines pollution in water bodies

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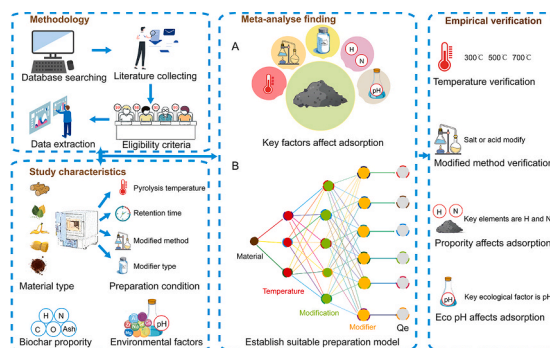
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HIGHLIGHTS

- First empirical research to validate the results of *meta-analysis*.
- *Meta-analysis* identified and predicted optimal conditions for biochar preparation.
- Adsorption capacity peaked at 637.71 mg/g, the highest value ever reported.
- Offers a new strategy for customised biochar preparation with specific functions.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Adsorption enhancement
Model validation
Modification optimisation
Simulation

ABSTRACT

The rapid increase in tetracycline pollution from intensive aquaculture necessitates urgent action to reduce tetracycline levels in water bodies, as its stability promotes antibiotic resistance, disrupts aquatic ecosystems, and poses health risks to humans through bioaccumulation. Current treatment methods like ozone oxidation are often costly or ineffective. Biochar, recognised for its cost-effectiveness and adsorption capacity, provides a promising solution. Through a *meta-analysis* of 110 research articles integrating 537 observations focused on plant-based biomass, this study identified the following: 1) key factors influencing biochar's adsorption of tetracyclines include preparation processes and environmental conditions; 2) higher pyrolysis temperatures can boost adsorption capacity, especially above 600 °C; 3) pre-treatment using acids or salts is more effective than other modifiers. Additionally, a linear mixed-effects model predicted that specific pyrolysis temperatures combined with acid or salt modification would produce biochar with superior performance. Verification experiments with pine, poplar, and straw from wheat, rice, and corn modified with salts (CaCl₂, ZnCl₂, MgSO₄, AlCl₃, FeCl₃, K₂FeO₄) and acids (H₃PO₄, H₂SO₄, HNO₃) revealed significantly increased adsorption capacities, as predicted by the linear mixed-effects model. The adsorption capacity of biochar prepared from biomass modified with 1 M H₂SO₄ for 24 h and pyrolysed at 700 °C can reach 637.71 mg/g, and the biochar exhibits enhanced adsorption capacity compared with those previously reported. Results of this study demonstrate the feasibility and accuracy

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of using *meta*-analysis to predict the performance of biochar in the efficient adsorption of tetracycline from water and provide a robust scientific framework for customised biochar preparation with specific functions.

1. Introduction

The rapid development of intensive agriculture, especially aquaculture, has led to excessive antibiotic use, with tetracyclines (TCs) being particularly problematic due to their broad-spectrum antibacterial activity and potential action as growth promoters (Scaria et al., 2021). Due to the poor metabolic absorption of TCs, over 75% is excreted unmetabolised or only partially metabolised into aquatic environments (Dai et al., 2020), disrupting ecosystems, fostering antibiotic-resistant bacteria (ARB) (Chopra, 2002; Xu et al., 2021), and inducing genotoxic effects on marine organisms and plants (Carballo et al., 2022; Zhou et al., 2019). Moreover, residual TCs can accumulate in fish or meat products, accelerating the growth of ARB in commensal organisms and causing harmful effects in humans (Nhung et al., 2017). Therefore, developing methods of alleviating TCs pollution is crucial for mitigating ecological risks.

Currently, TCs are mainly removed using adsorption techniques. Other removal processes include plasma treatment (Shen et al., 2024), Fenton oxidation (Zhang et al., 2019), electrochemical oxidation (Chen et al., 2024), and photocatalysis (Wu et al., 2020) among others, but face the drawbacks of high operational costs, limited removal efficiency, or harmful by-products (Bhagat et al., 2020). For example, membrane separation retains 92% of TCs but generates highly concentrated waste, causing secondary pollution (Li et al., 2004). Ozone oxidation can fully degrade TCs but requires significant equipment and energy investment (Iakovides et al., 2019). Photocatalysis is environmentally friendly but faces challenges in catalyst recovery (Wang et al., 2020). Thus, it is essential to find cost-effective and efficient adsorption methods without harmful by-products. Compared with adsorption materials such as activated carbon, zeolites, and clay, biochar has significant advantages in energy consumption, renovation, and recyclability, making it a promising option for TCs removal (Tan et al., 2017).

Biochar, a carbon-rich material produced from biomass pyrolysis, is a cost-effective option for antibiotic remediation due to its exceptional adsorption capacity, particularly for TCs. Sourced from agricultural waste, it exemplifies a 'waste-to-resource' approach. Its porous structure and its functional groups enhance the adsorption capacity, offering practical water remediation solutions. Recent studies underscore biochar's potential for sustainable TCs removal. Liu et al. (2025a) achieved 52.90 mg/g TC adsorption with grape-based biochar while Li et al. (2025) reported 310.70 mg/g adsorption with bamboo-based biochar. However, the adsorption performance depends on a range of factors including pyrolysis and modification conditions. For instance, raising the pyrolysis temperature of cow manure-derived biochar from 300 °C to 700 °C can double the adsorption capacity of the biochar (Zhang et al., 2019). Additionally, incorporating metal ions such as Ca, Mg, and Al into biochar can increase the adsorption capacity (Liu and Fan, 2023). These observations illustrate the complexity of optimising biochar, as varying aspects of the preparation conditions affect its physicochemical properties and adsorption capacity. Therefore, tailoring biochar for specific scenarios is crucial for maximising TCs adsorption efficiency.

Current studies often focus on isolated variables in the preparation process without considering multifactorial interactions, leading to sub-optimal outcomes and limited generalisability. By integrating data across studies, *meta*-analysis, a statistical technique that combines and analyses the results of multiple independent studies to reach an overall conclusion (Jones et al., 2008), offers a robust framework to address these gaps, enabling a comprehensive understanding of how preparation methods influence biochar properties and adsorption capacity. *Meta*-analysis has been widely applied in environmental remediation research. For example, Carpio et al. (2018) used it to study the link

between microbial diversity and heavy metal remediation, clarifying that microbial consortia effectively reduce ecological pollution. A *meta*-analysis by Liu et al. (2025b) highlighted that microorganisms effectively degrade and optimise the breakdown of organic pollutants. Despite their widespread application in environmental remediation research, the results of *meta*-analyses have received limited experimental verification.

Taking TCs adsorption by customised biochar as an example, this study verified the feasibility and accuracy of *meta*-analysis in systematically evaluating the relationships between biochar preparation conditions and biochar adsorption capacity. Initially, key factors influencing the adsorption of TCs by biochar were analysed and screened using a *meta*-analysis based on 537 observations. Subsequently, based on the analytical results, a linear mixed-effects model was developed to predict the optimal preparation conditions for maximising the adsorption capacity of biochar. Biochar was then prepared under these conditions to assess the predictive effectiveness of *meta*-analysis in practical applications. The environmental factors affecting the adsorption of TCs by biochar were also analysed. This study aims to provide practical insights for customising biochar tailored to specific environmental conditions, thus contributing to advancing sustainable aquaculture practices, reducing ecological risks, and promoting environmental protection through innovative water remediation strategies.

2. Material and methods

2.1. Data collection

Articles published between 1 January 2000 and 1 May 2024 were retrieved from the Web of Science database (<https://www.webofscience.com/wos/>) using specific keywords ([biochar OR bio-char OR charcoal] AND [Tetracyclines OR Oxytetracycline OR Chlortetracycline OR Doxycycline OR Tetracycline]). The decision to exclude aquatic-related keywords was made in order to broaden the literature scope, with the intention of filtering out irrelevant literature manually thereafter. Initially, 1,124 articles were retrieved, with 110 meeting the following selection criteria: 1. Clear records of biochar production process (raw materials, preparation, modification) and biochar properties; 2. Data including the parameters of adsorption experiments (biochar dosage, TCs solution concentration, volume) and measured TCs adsorption data (mean, SD/SE); and 3. Studies that used plant materials rather than materials like sludge or animal manure, and focused specifically on TCs adsorption in water bodies, excluding review articles (see [Supplementary Materials](#)). The extracted data encompassed raw material types, biochar preparation procedure (pyrolysis temperature, retention time), modification method (no modification, biomass modification, biochar modification, or further detailed description), biochar properties (C%, H%, O%, N%, ash content), and environmental factors (pH; ion type and concentration). The detailed definitions of these parameters can be found in [Supplementary Materials](#). To predict the TCs adsorption capacity (Q_e , mg/g) of biochar samples and evaluate the diverse influences of biochar properties on TCs adsorption, a comprehensive investigation involving the factors specified above was conducted by *meta*-analysis. Data from figures were extracted using the WebPlotDigitizer online software (Marin et al., 2017; Wang et al., 2024). The same researcher extracted the data multiple times and checked the consistency of the data after each extraction.

2.2. Meta-analysis and data analysis

Meta-analysis was conducted to quantify the average effect of

biochars, grouped in terms of various factors, using the natural log-transformed response ratio (lnRR) as the metric. The effect sizes were weighted by the inverse of the pooled variance (v). Heterogeneity was evaluated using the Cochrane Q-test and I^2 index, revealing significant heterogeneity ($I^2 > 75\%$). Subgroups were created accordingly, and a random-effects model with restricted maximum likelihood estimation was used to account for within- and between-study variances. Heterogeneity metrics were computed using the 'metafor' package v4.6–0 (Viechtbauer, 2010) in R, with Hartung–Knapp adjustment for small-study effects (Hartung, 1999). Linear mixed-effect models for material type, pyrolysis temperature, and modification method were constructed using the R packages 'broom' v1.0.6 (Robinson et al., 2024) and 'lme4' v1.1–35.5 (Bates et al., 2015). Factor relationships were visualised with the 'pheatmap' package v1.0.12 (Kolde, 2019) and 'ggplot2' v3.5.1 (Ginestet, 2011). Missing values were handled by data removal, category conversion, or multiple imputations via the 'mice' package (van Buuren and Groothuis-Oudshoorn, 2011).

Publication bias assessments were performed using Rosenberg's method, with Rosenberg's fail-safe number calculated accordingly (Rosenberg, 2005). A fail-safe number is considered robust if it exceeds $5n + 10$, where n represents the original number of studies. For all the studies in the meta-analysis (see Supplementary Materials), the values of $5n + 10$ are less than the fail-safe number. Therefore, the collected data are free of bias. The specific formulas and calculation methods can be found in Supplementary Material 1.2.

2.3. Biochar preparation and characterisation

Biochar was prepared under conditions based on the meta-analysis, which identified biomass modification using salts (CaCl_2 , ZnCl_2 , MgSO_4 , AlCl_3 , FeCl_3 , K_2FeO_4) or acids (H_3PO_4 , H_2SO_4) as effective. The materials described in Supplementary Material 1.3 were used for raw and modified biochar production, and detailed preparation methods are described in Supplementary Material 1.4. The biochars prepared from biomass modified with H_3PO_4 , H_2SO_4 , HNO_3 , CaCl_2 , MgSO_4 , ZnCl_2 , and FeCl_3 are respectively denoted as PXBC, SXBC, NXBC, CaXBC, MgXBC, ZnXBC, and FeXBC, where X represents R (RBC), W (WBC), C (CBC), Po (PoBC), or P (PBC). The numbers after the biochar names represent the pyrolysis temperature. The C, N, and H contents of raw or modified biochars were analysed using an elemental analyser (Vario EL cube, Germany). The pH of zero point charge (pHPZC) values for the raw and modified biochars were estimated by the pH drift method (Qin et al., 2022).

2.4. Adsorption experiments

Tetracycline hydrochloride (TCH) was selected as a model TC. TCH was dissolved in deionised water with natural pH. The concentration of TCH was quantitatively measured using a UV-visible spectrophotometer (UV2400, China) at 357 nm, establishing a calibration curve based on different TCH concentrations (12.5, 25, 50, 100, 200 mg/L) (see Supplementary Materials). Adsorption experiments were conducted in 14 mL centrifuge tubes containing 10 mL of TCH solution (200 mg/L) with 0.01 g of biochar, following the method of Li et al. (2021) with slight modifications. The tubes were placed in a thermostatic oscillator (ZQZY-78A, China) at 30 °C with a shaking speed of 200 rpm. For kinetic experiments, samples were collected at 0, 1, 3, 5, 7, 9, 12, 24, and 48 h. For isotherm experiments, biochar was exposed to TCH solutions with concentrations of 12.5, 25, 50, 100, 200, 400, 800, and 1,600 mg/L for 24 h at 30 °C. All experiments were performed in triplicate. After adsorption, the suspensions were centrifuged and filtered using a 0.22 µm syringe filter (PES), and residual TCH concentrations were measured at 357 nm.

Due to the significant surface heterogeneity of the material, the pseudo-first-order model was used, instead of the pseudo-second-order model, for fitting the kinetic data. The adsorption isotherms were

determined using the Freundlich, Langmuir, and Sips models. The detailed process of the adsorption experiments and the specific formulas for the adsorption models are detailed in Supplementary Materials.

To investigate the effect of solution pH on the adsorption process, the pHPZC of the prepared biochar samples was first determined using the pH drift method described in 2.3. Subsequently, batch experiments were conducted at various pH levels ranging from 2 to 10 to examine the adsorption capacity of biochar (0.1 g) for TCH solution (200 mg/L, 100 mL, 30 °C, 24 h) under different pH conditions. The pH of the TCH solution (200 mg/L) was adjusted using 0.1 M HCl or NaOH with a slight modification of the method described by Qin et al. (2022).

3. Results and discussions

3.1. Meta-analysis of factors affecting biochar adsorption

3.1.1. Key factors affecting adsorption of tetracycline

Identifying key factors in biochar preparation is crucial for optimising its adsorption capacity. The key factors affecting TCs adsorption include pyrolysis temperature, modification method, N and H content, and environmental pH (solution pH) (Fig. 1A). Specifically, pyrolysis temperature, modification method, and environmental pH are highly significant ($p < 0.001$), aligning with literature emphasising their roles in enhancing adsorption capacity: higher pyrolysis temperatures can increase biochar surface area and porosity, while modification methods introduce functional groups that enhance adsorption (Zeng et al., 2018), while solution pH can affect the biochar's polarity, surface charge, and ion exchange capacity, further influencing biochar adsorption performance (Zhang et al., 2024). Conversely, the N and H contents influence adsorption but with weaker significance ($p = 0.006$, $p = 0.016$, respectively). Potential mechanisms of their effects include low hydrogen content reducing the H/C ratio, which in turn increases biochar aromaticity and the number of adsorption sites (Zhang et al., 2023). The N and H contents also affect the surface charge and can influence electrostatic adsorption by altering functional groups' pH or protonation state (Hsu et al., 2019). Factors found not to be significant include material type, retention time, C and O content, ash content, and ionic solubility. However, these factors may exhibit heterogeneity among subcategories, leading to their non-significant overall effects. Thus, significant factors were analysed in the meta-analysis, while non-significant factors were examined for subcategory effects. Particle sizes could not be verified due to the crushing of raw materials during pre-preparation.

To enhance the adsorption capacity of biochar, it is essential to evaluate key preparation factors and their potential interactive effects. For this purpose, Pearson's correlation coefficient was employed to analyse the relationships between the effect values and these factors (Fig. 1B). Most factors (all except 'Wood and Shell', 'Biochar Alkali', 'Biochar Pyrolysis', 'Biochar Salt', and '200–400 °C') showed negative correlations with lnRR (correlation coefficients < 0 , $p < 0.05$) (see Supplementary Materials), suggesting that preparation conditions can enhance adsorption capacity. The lnRR values exhibited weak correlations among all preparation factors (absolute correlation coefficients < 0.3), revealing that the complex interplay of multiple factors influences the adsorption performance. Additionally, element pairs showed weak interactions (absolute correlation coefficients < 0.3), illuminating that changes in individual factors do not significantly affect adsorption characteristics. Thus, a holistic approach that considers the synergistic effects of various preparation parameters is necessary to achieve optimal performance.

3.1.2. Influence of biomass source on biochar adsorption capacity

Biomass, a composite material of biological, organic, and inorganic substances from living organisms, varies significantly in chemical composition. These components' behaviours during pyrolysis determine biochar's chemical structure and properties (Seow et al., 2022).

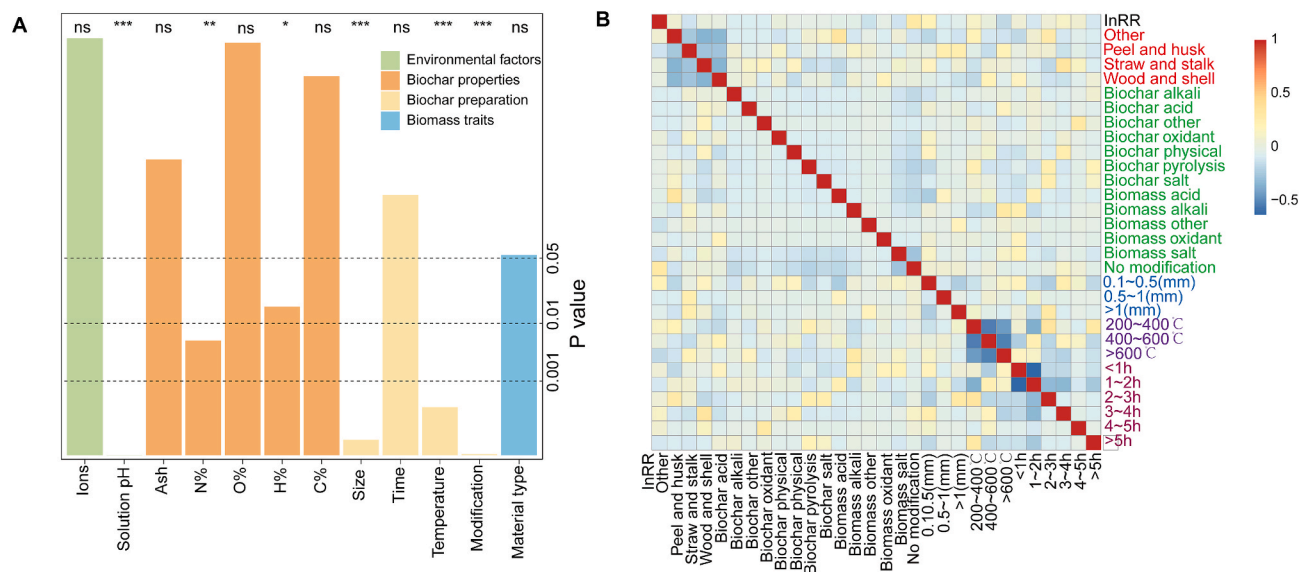


Fig. 1. Factors affecting biochar adsorption. A. Various factors impact on TCs adsorption by biochar, with significance levels indicated by * ($0.01 < p < 0.05$), ** ($p < 0.01$), *** ($p < 0.001$), and 'ns' ($p > 0.05$) (The same as below). B. Pearson correlation matrix between InRR and preparation conditions: red (material type), green (modification method), blue (particle size), purple (pyrolysis temperature), magenta (retention time).

Conventional understanding indicates that the adsorption capacity of biochar is influenced by the different functional groups derived from various biomass types (Deng et al., 2016; Tomczyk et al., 2020). However, biomass type is not a critical factor influencing biochar's adsorption capacity for TCs in water bodies according to the *meta*-analysis (Fig. 1A). For different materials made from raw biochar, the density peaks of the adsorption amount are similar, indicating relatively consistent adsorption capacity. Data analysis revealed that the pore volumes of the materials were mostly less than $0.3 \text{ cm}^3/\text{g}$ and had similar the average pore diameter, indicating that the materials had largely similar characteristics that resulted in comparable (and limited) adsorption performance (see [Supplementary Materials](#)). Despite the introduction of different functional groups (Wang and Wang, 2019), the adsorption potentials of the biochars modified with 'Leaf', 'Peel and Husk', and 'Wood and Shell' were similar to each other, showing comparable capacities and low variability (see [Supplementary Materials](#)).

The materials were grouped to analyse the differences between each group. Although significant differences exist among the categories, the effect sizes are relatively small: the InRR values for 'Wood and shell' (-0.67), 'Straw and stalk' (-0.70), 'Peel and husk' (-0.92), 'Other' (-0.63), and 'Leaf' (-0.60) have overlapping 95% confidence intervals (see [Supplementary Materials](#)). The overlap in effect-size ranges suggests no clear superiority among the material categories. Thus, despite variations in feedstock, the different biochar types show comparable specific adsorption capacity, suggesting that the feedstock's influence on biochar's adsorption capacity may be less significant than previously believed.

3.1.3. Influence of modification and pyrolysis temperature on adsorption capacity

Modification methods – chemical, physical, and biological – affect adsorption capacity by introducing heteroatoms and altering pore structure (Lan et al., 2024). Modifications can be divided into pre-treatment or post-treatment (corresponding to biomass or biochar modification in this study). *Meta*-analysis revealed that biochar modification generally enhances adsorption less strongly than biomass modification does, possibly due to the greater difficulty of functional-group introduction into biochar because of its stable structure (Fig. 2A) (Tomczyk et al., 2020). Huang et al. (2023) found that pre-treatment enhanced surface area, pore volume, and functional groups

significantly more than post-treatment did. Thus, biomass modification may enhance the pollutant adsorption capacity of the material more effectively than biochar modification. The approaches for modifying biomass and biochar were subdivided to explore the variations among materials treated by different modification methods (Fig. 2B). It was found that acid or salt modification in pre-treatment has promising effects, while alkaline or oxidant modification in post-treatment can also enhance TCs adsorption.

For raw biochar, the adsorption capacity is significantly influenced by the pyrolysis temperature, which determines its physicochemical properties and structure, affecting its elemental composition, pore structure, surface area, and functional groups (Tag et al., 2016). Based on the degradation temperatures of hemicellulose, cellulose, and lignin, pyrolysis temperatures can be categorised into three ranges: 200–400 °C, 400–600 °C, and above 600 °C (Amalina et al., 2022; Sakhiya et al., 2020). For raw biochar, the comprehensive *meta*-analysis (see [Supplementary Materials](#)) and modelling results (Fig. 2C) showed that higher pyrolysis temperatures enhance TCs adsorption, with temperatures above 600 °C being optimal, followed by the 400–600 °C range. For modified biochar, both temperature and modifications influenced the adsorption capacity. However, few studies have explored the interactions between temperature and modifications. Thus, this study established a mixed model that correlated specific modification methods, materials, and pyrolysis temperature to predict adsorption capacity.

As can be seen in Fig. 2C, optimal adsorption can occur at different temperatures depending on the modification methods. For biomass modification, the modification effect generally increases slightly with temperature, whereas for biochar modification, the modification effect is less significant with increasing temperature (Fig. 2C). Due to insufficient data on other modifier, acid or salt modification in pre-treatment was selected for further analysis. Salts and acids were categorised by ion type and chemical properties based on the collected dataset, excluding less frequently used salts and acids. Despite some missing data points, discernible trends emerged. At 200–400 °C, biomass modified with Zn^{2+} , Mg^{2+} , and Al^{3+} salts demonstrated enhanced adsorption capacity, although some ranges of data for Mg^{2+} , and Al^{3+} were lacking. The efficacy of FeO_4^{2-} salt modification at 200–400 °C or > 600 °C surpassed that at 400–600 °C. The effect was optimal for iron- and calcium-modified biomass at > 600 °C, although data for Ca^{2+} at

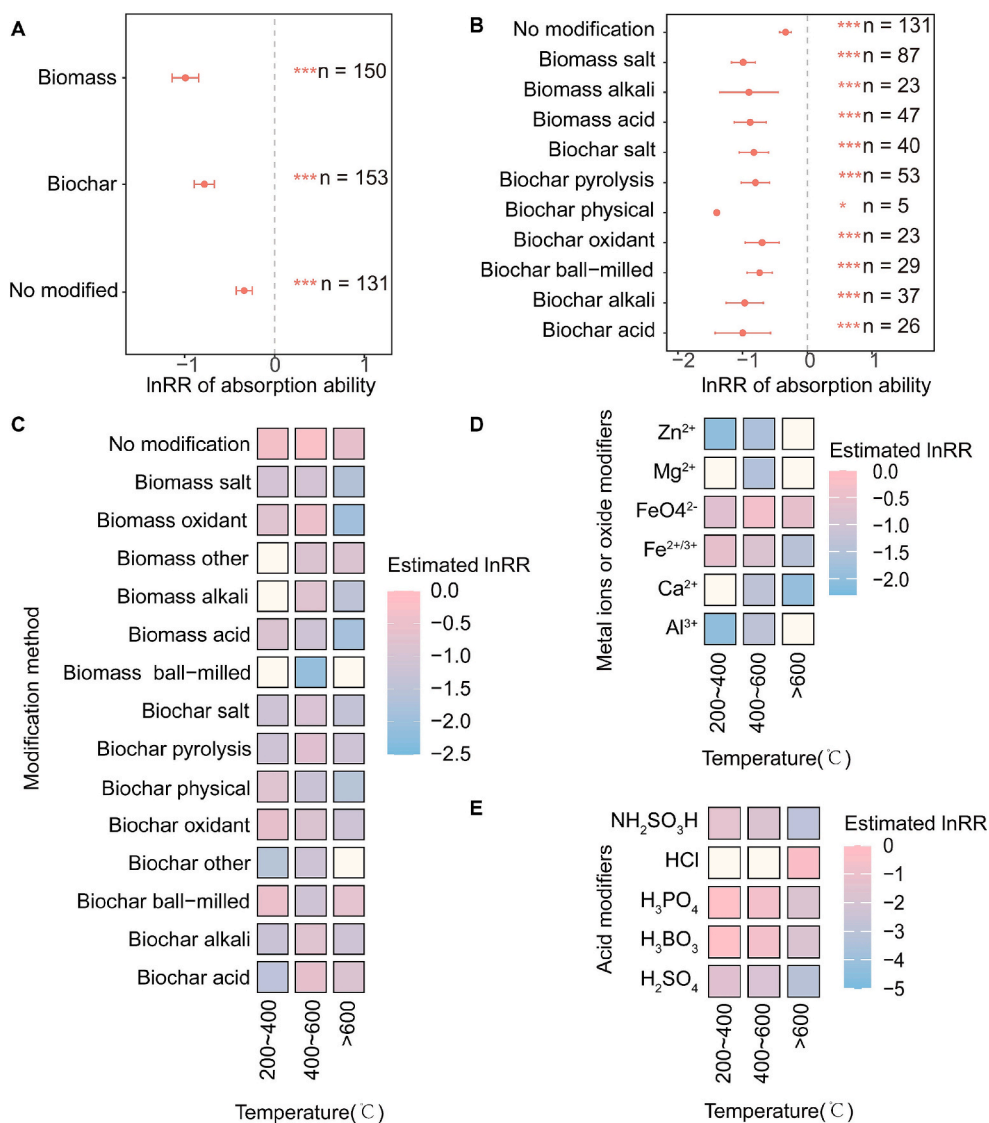


Fig. 2. Meta-analysis of effect of biochar modifications on TCs adsorption. (A) Effects of modification method types, including un-modified, pre-treatment ('Biomass'), and post-treatment ('Biochar'), on adsorption capacity values. (B) Effects of specific modifiers on adsorption capacity values after pre-treatment and post-treatment. Simulation using linear mixed-effects models of modification methods and pyrolysis temperatures (C), salt types (D), and acid types (E). Adsorption capacity is indicated by colour: sky blue (high) and pink (low); ivory indicates missing or invalid data.

200–400 °C were lacking (Fig. 2D). For acid modification, sulfamic acid and H₃BO₃ modification resulted in the highest adsorption capacity at > 600 °C, while HCl modification had moderate effects across all temperatures. The effectiveness of modification with H₃PO₄ and H₂SO₄ increased with temperature, peaking at > 600 °C (Fig. 2E). Overall, for most acid and salt modifications, the probability of improving the adsorption capacity of biochar increases when the pyrolysis temperature exceeds 600 °C.

The meta-analysis results indicated that pyrolysis retention time is not a key factor affecting biochar adsorption capacity (Fig. 1A). Biochars were divided into four groups by retention time: 0–1 h, 1–2 h, 2–3 h, and more than 3 h. The first three groups showed similar effect sizes within 3 h retention time (0–1 h: -0.71, 1–2 h: -0.75, 2–3 h: -0.72) (see Supplementary Materials), suggesting that short-duration pyrolysis is sufficient to form biochar with favourable adsorption properties. This can be attributed to the minor impact of retention time on biochar structure, as rapid pyrolysis under high-temperature conditions quickly stabilises biochar properties (Arslanoğlu et al., 2023). Thus, a retention time of 1 h is recommended for energy conservation in practice.

3.1.4. Influence of elemental composition on biochar properties and adsorption capacity

Limited research has explored the relationship between the elemental composition of biochar and its TCs adsorption performance. Based on the meta-analysis results, the H and N contents in biochar significantly influence its adsorption capacity for TCs while those of C and O do not. In further analysis, biochars were categorised into three levels by adsorption capacity: high (> 300 mg/g), medium (100–300 mg/g), and low (< 100 mg/g) (see Supplementary Materials). Meta-analysis showed that biochars with adsorption capacities exceeding 100 mg/g are more likely to have an H content within 0–3% (total range: 0–8%) and an N content within 0–2% (total range: 0–5%) (see Supplementary Materials). In contrast, the C contents were mainly greater than 40%, and the O contents ranged from 0% to 40%, with no specific range corresponding to significantly higher adsorption capacities (see Supplementary Materials). Interestingly, in the preparation process, the modification methods significantly affected the content of C and O, while for the H and N contents, the pyrolysis temperature, retention time, and material type were important (see Supplementary Materials). In brief, the adsorption performance of biochar is most likely to be

optimal when the H content is between 0% and 3% and the N content is between 0% and 2%.

3.1.5. Influence of environmental pH on biochar adsorption capacity

Research indicates that solution pH, which is closely associated with the functional groups on the biochar surface, significantly influences the adsorption of contaminants onto biochar (Ambaye et al., 2021). Meanwhile, solution pH variations alter the surface charge and ionisation of biochar, affecting the contaminant removal capacity (Zhang et al., 2013). Consistent with the literature, *meta*-analysis showed that environmental pH is a crucial factor affecting TCs adsorption onto biochar (Fig. 1A). Subgroup analyses revealed that the adsorption performance is better when solution pH at 1, 3, 5, 7, 9, and 11 than at pH 2, 4, 6, 8, and 10 (see Supplementary Materials). Further analysis showed that biochar's adsorption capacity tends to increase slightly with solution rising pH from 1 to 7 but decreases as pH increases from 7 to 12. This biphasic response to pH indicates that a neutral solution pH range is optimal for biochar application in remediating water contaminated with organic pollutants. However, biochar may also demonstrate good adsorption of pollutants at other solution pH ranges, as the differences between these values are very small.

3.2. Experimental validation of conditions for predictive meta-analysis in biochar production

3.2.1. Meta-analysis of the predictive effectiveness for material type

To test the effectiveness of the *meta*-analysis-predicted conditions for biochar preparation, three types of straw (rice, wheat, and corn), two types of wood (pine and poplar), citrus peel, and maple leaves were selected (the 'Other' group was excluded due to the complexity of the types of materials in this group). The measured adsorption capacities of biochar produced at three pyrolysis temperatures – i.e., 300 °C, 500 °C, and 700 °C, selected from the temperature ranges used in the *meta*-analysis (200–400 °C, 400–600 °C, and > 600 °C) – were used to validate the adsorption performance of biochar produced from different biomass sources.

The results revealed that the adsorption capacities varied significantly across pyrolysis temperatures, while the influence of biomass types was less pronounced. At 300 °C, rice straw (60.18 mg/g), wheat straw (57.41 mg/g), and corn straw (57.94 mg/g) exhibited comparable capacities, while those of pine (57.94 mg/g) and poplar (54.33 mg/g) were slightly lower. Citrus peel demonstrated the highest capacity (66.46 mg/g), and maple leaves the lowest (54.65 mg/g) (see Supplementary Materials). At 500 °C, corn straw achieved the highest capacity (75.64 mg/g), followed by wheat straw (65.91 mg/g), maple leaves (59.12 mg/g), and rice straw (51.46 mg/g). Pine (64.44 mg/g) and poplar (60.71 mg/g) had similar adsorption capacity, whereas citrus peel had a lower capacity (39.33 mg/g) (see Supplementary Materials). At 700 °C, corn straw had the highest capacity (85.18 mg/g), followed by rice straw (82.42 mg/g) and wheat straw (70.82 mg/g). Poplar (83.48 mg/g) and pine (73.48 mg/g) were comparable. Citrus peel and maple leaves had almost identical capacities (71.35 mg/g each) (see Supplementary Materials). These findings indicate that while there are some distinctions in adsorption capacities among materials, the overall differences are not pronounced, as indicated by the overlap in performance ratings. Thus, specific biomass selection is not essential for enhancing biochar adsorption. These results broaden the scope of raw materials for biochar production for diverse environmental application options, potentially improving resource efficiency and economic viability.

3.2.2. Effect of salt or acid pre-treatment and pyrolysis temperature on adsorption capacity of biochars

The *meta*-analysis indicated that retention time is not a critical factor influencing the adsorption capacity of biochar during its preparation. Empirical experiments with straw and wood materials prepared at a

pyrolysis temperature of 700 °C revealed minimal and inconsistent effects of retention time (1–3 h) on adsorption capacity, which verified the *meta*-analysis-predicted results (see Supplementary Materials). This can be attributed to the minor impact of retention time on biochar structure, as rapid pyrolysis under high-temperature conditions quickly stabilises biochar properties (Chen et al., 2023; Osman et al., 2022). Thus, a retention time of 1 h is sufficient to meet the requirements for biochar production.

The *meta*-analysis and modelling results accurately predicted that raw biochar's adsorption capacity would increase with higher pyrolysis temperatures, as described in the 3.2.1 section. Based on material availability and cost (wheat straw, rice straw and corn stalk: 16 RMB/kg, pine wood: 12 RMB/kg, poplar wood: 22 RMB/kg, orange peel: 59.6 RMB/kg, ivy leaf: 29.8 RMB/kg; data from Taobao [<https://www.taobao.com/>]), wood and straw materials were selected for the verification experiment using modified biochar. Fig. 3 illustrates the adsorption capacity of various salt- or acid-modified, biomass-derived biochars for TCH prepared at temperatures of 300 °C, 500 °C, and 700 °C, respectively. With salt or acid concentrations of 0.1, 0.25, 0.5, and 1 M, the corresponding variation in the adsorption capacity of biochar produced from modified biomass was relatively small (see Supplementary Materials). Thus, a salt concentration of 0.25 M and an acid concentration of 1 M were chosen for biomass modification to mitigate the adverse effects of high concentrations of salts or acids on the physical properties.

The activation trends of biomass varied with salt and temperature. Interestingly, whether at 300 °C, 500 °C, or 700 °C, certain salts consistently activated the biomass materials more effectively than others, regardless of material type. For instance, at 300 °C, FeCl₃ demonstrated the greatest activation effect on rice straw (TCH adsorption: 132.05 mg/g), followed by CaCl₂ (129.22 mg/g), while ZnCl₂ showed the weakest activation (75.07 mg/g). K₂FeO₄ exhibited a moderate activation effect (101.24 mg/g) (Fig. 3A). At 500 °C, ZnCl₂ exerted the greatest activation effect on rice straw (119.23 mg/g), followed by MgSO₄ (139.65 mg/g), with FeCl₃ showing the weakest activation (72.20 mg/g) (Fig. 3B). At 700 °C, the activation effect of ZnCl₂ on rice straw (113.59 mg/g) diverged from those of other salts (Fig. 3C). All the tested salt modifications exhibited similar trends in the other two straw and wood materials (see Supplementary Materials). Similarly, the acids used in the validation experiments – H₃PO₄, H₂SO₄, and HNO₃ – also showed this trend; specific acids consistently exhibited significantly higher activation efficacy on biomass substrates across different material types. The activation levels of straw materials modified with H₂SO₄ and HNO₃, but not H₃PO₄, increased with temperature and reached maximum values at 700 °C (Fig. 3D-F). In contrast, due to the corrosive nature of H₃PO₄, which increases material porosity during preparation, temperature had no significant impact on the activation effect of H₃PO₄ modification (Jiang et al., 2024). Although the adsorption capacities of different materials with the same modifiers were generally similar, wheat straw biochar demonstrated the highest capacity in most cases (Fig. 3, and see Supplementary Materials). Thus, wheat straw was chosen for the subsequent experiments.

The predictions of the model established in this study are broadly consistent with most of the actual outcomes (Figs. 3-4). For example, modification with most salts and acids improves the adsorption capacity of biochar pyrolysed at 700 °C, with FeCl₃, AlCl₃, CaCl₂, MgSO₄, H₂SO₄, and HNO₃ achieving the best performance, aligning with the observation that a higher pyrolysis temperature (> 600 °C) enhances adsorption capacity. However, discrepancies are observed for some cases, like zinc salts, where the model predicts an optimal pyrolysis temperature of 300 °C, whereas the actual optimal temperature is 500 °C. Similarly, iron salts performed best for biochars pyrolysed at 300 °C, which contradicts the model's predictions. These inconsistencies may arise from the limited dataset used for analysis.

Despite some missing data, the model successfully identified the optimal pyrolysis temperatures for most salts: 300 °C for Al³⁺ salts, 700 °C for Ca²⁺ salts, 300 °C and 700 °C for FeO₄²⁻ salts, and 500 °C for

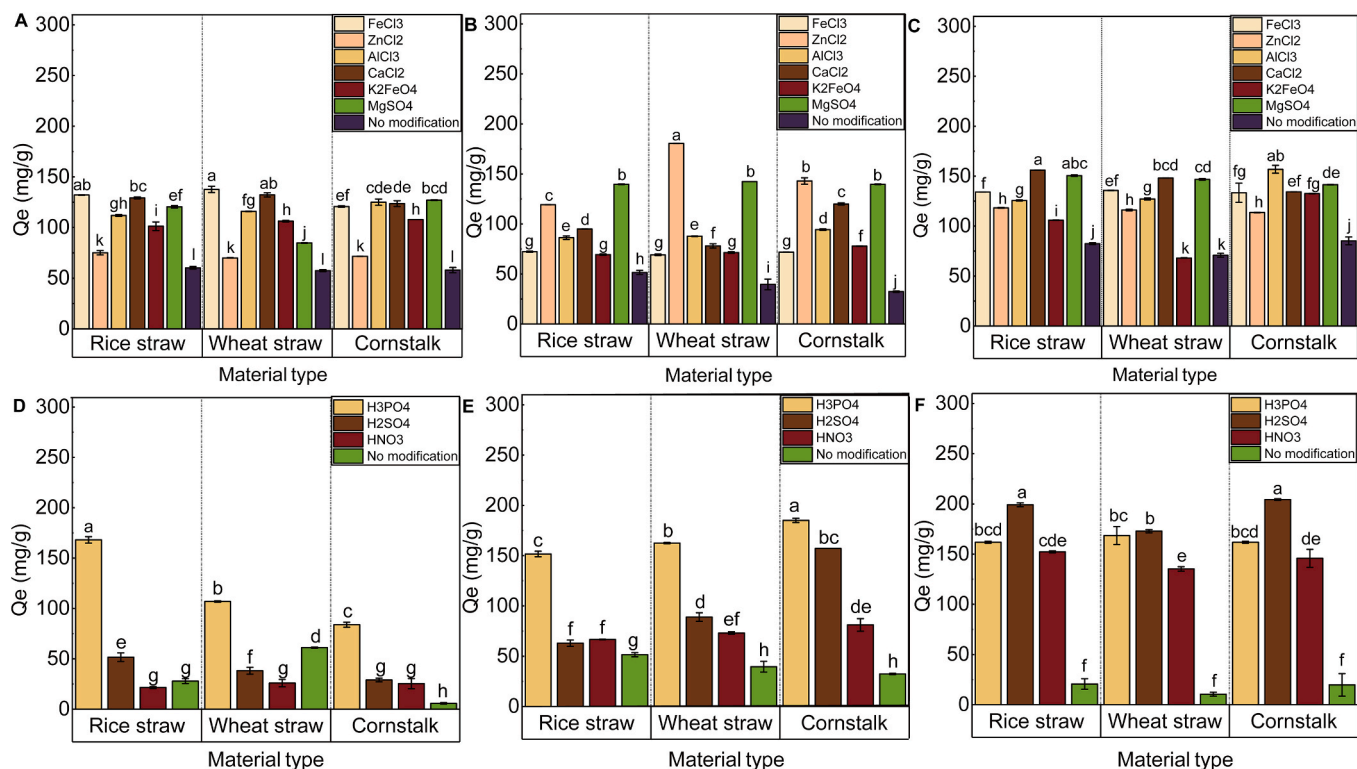


Fig. 3. Effects of various salt- or acid-modified biomasses on the adsorption of TCH by biochar. Adsorption efficiency of tetracycline on biochar derived from different salt-modified biomasses pyrolysed at 300 °C (A), 500 °C (B), and 700 °C (C). Adsorption efficiency of tetracycline on biochar derived from different acid-modified biomasses pyrolysed at 300 °C (D), 500 °C (E), and 700 °C (F).

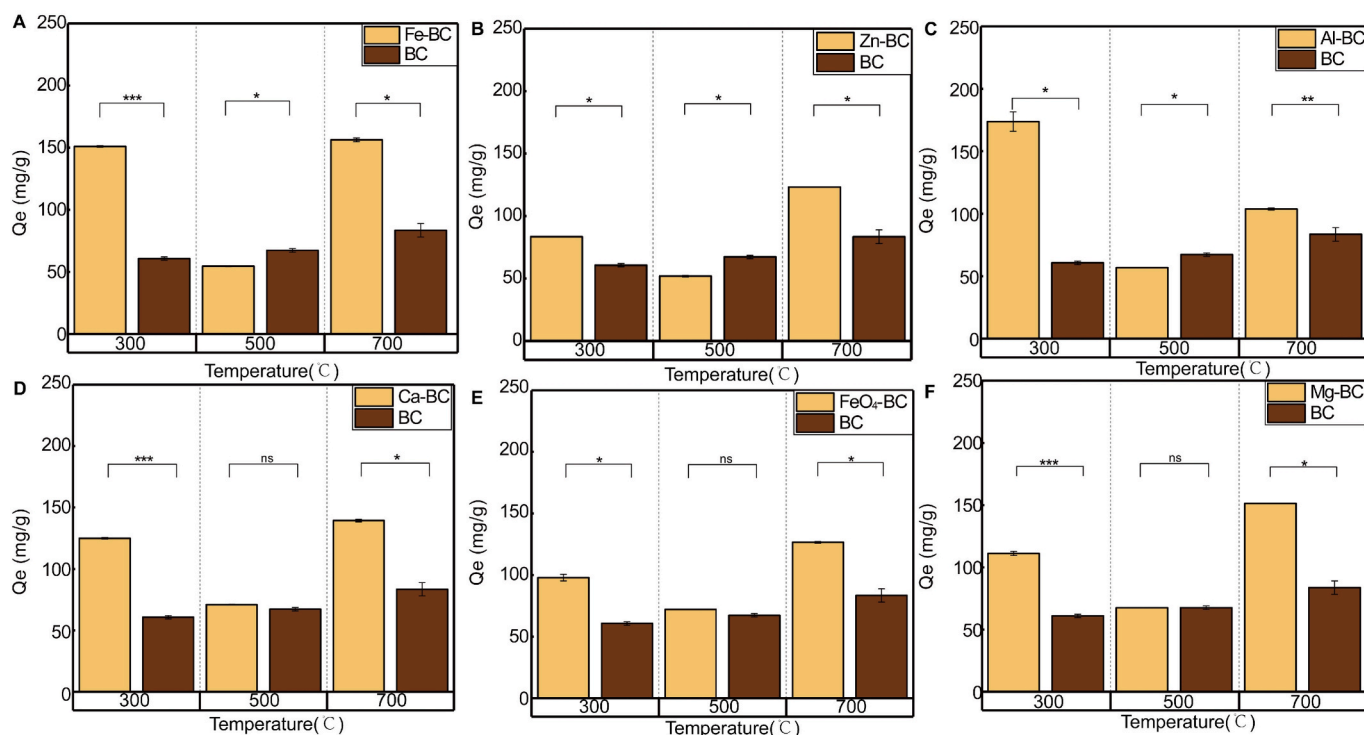


Fig. 4. Adsorption efficiency of TCH on biochar produced from FeCl₃-modified (A), ZnCl₂-modified (B), AlCl₃-modified (C), CaCl₂-modified (D), K₂FeO₄-modified (E), and MgSO₄-modified (F) wheat straw at 300 °C, 500 °C, and 700 °C.

Mg²⁺ salts. Meanwhile, under the optimised modification conditions, salt modification enhanced the biochar adsorption capacity from 2.16–46.6 mg/g to 74.1–122.1 mg/g (adsorption rate: 37–61%), while

acid modification further increased it to 172.9–204.4 mg/g (adsorption rate: 86.4–100%, TCH: 200 mg/L). These findings underscore the model’s utility in identifying optimal biochar production conditions and

guiding experiments to enhance biochar characteristics and adsorption capacity.

3.2.3. Effect of elemental composition on predicted adsorption efficiency of biochars

H₃PO₄-modified biochars (PWBC300, PWBC500, PWBC700), raw biochar (WBC500), and one salt-modified biochar (ZnWBC500) were selected to verify the *meta*-analysis of the effect of elemental composition on biochar adsorption capacity (see [Supplementary Materials](#)). High-adsorption biochars were found to have N and H contents of 0–2% and 0–3%, respectively, aligning with the *meta*-analysis predictions. In contrast, low-adsorption biochars like WBC500 have H contents exceeding 3%. Although both PWBC300 (59.27% C) and WBC500 (64.93% C) have C contents exceeding 40%, their adsorption capacities (168.12 mg/g and 51.46 mg/g, respectively) show significant differences, indicating that C content alone does not strongly correlate with adsorption capacity, consistent with the *meta*-analysis results. These findings confirm that N and H are the primary determinants of biochar adsorption performance.

3.2.4. Effect of environmental solution pH on predicted adsorption capacity of biochars

The solution pH level significantly influences the biochar adsorption process. TCH has three acid dissociation constants (pK_a values of 3.30, 7.68, and 9.69). The primary species of TCH are H₄TC⁺ at pH less than 3.3, H₃TC at pH 3.4–7.3, H₂TC⁻ at pH 7.3–9.4, and HTC²⁻ at pH > 9.4 (Croitoru et al., 2020). Various modified biochars pyrolysed at 700 °C were selected to study the effect of environmental pH on biochar adsorption. NWBC700, PWBC700, SWBC700, ZnWBC700, MgWBC700, and CaWBC700 exhibit pHPZC values of 5.74, 6, 9.56, 7.37, 9.24, and around 10, respectively (see [Supplementary Materials](#)).

At pH < 3.3, under acidic conditions, H₄TC⁺ and all biochars possess positive surface charges, resulting in strong electrostatic repulsion and consequently lower adsorption efficiency for all biochar types. In the 3.3 < pH < pHPZC range, TCH is predominantly neutral (H₃TC), and the biochar surfaces remain positively charged below their respective pHPZC values. The elimination of electrostatic repulsion allows non-electrostatic interactions (π - π electron donor-acceptor [EDA], hydrogen bonding, pore-filling) to dominate adsorption (Ahmed et al., 2018). As for PWBC700 and NWBC700, adsorption efficiency increases significantly within their respective operational ranges, driven by these non-electrostatic forces. For SWBC700 (pHPZC 9.56), the adsorption capacity remains relatively stable at pH 3.3–7.3. When pH increases further (7.3 < pH < 9.56), TCH transforms to H₂TC⁻ while the SWBC700 surface charge weakens but remains near neutral or weakly positive. Electrostatic repulsion is minimal or absent, allowing π - π EDA and pore-filling to maintain stable adsorption. For CaWBC700, MgWBC700, and ZnWBC700, a decrease in adsorption capacity (slight or significant) is observed, attributable to specific precipitation or competitive adsorption mechanisms. When the pH is around 6 for Ca²⁺, 8 for Mg²⁺, and within the range of 5 to 7 for Zn²⁺, precipitates or hydrolysis products will form. For CaWBC700 in non-degassed solutions, CaCO₃ precipitates readily form around pH 6 due to dissolved CO₂/Ca²⁺, coating surfaces and blocking pores (Xie et al., 2018). Mg²⁺ forms Mg(OH)₂ colloids/precipitates around pH 8, causing less severe blocking due to higher solubility (Phillips et al., 1977). Zn²⁺ forms ZnOH⁺ (and Zn(OH)₂) between pH 5–7 with a positive charge and can compete with TCH for adsorption sites on the biochar. These processes reduce adsorption capacity in their respective pH ranges by covering active sites, blocking pores, or direct competition (Fedorova et al., 2018).

When pH > pHPZC, both TCH species (H₂TC⁻/HTC²⁻) and the biochar surfaces carry negative charges, leading to electrostatic repulsion. However, adsorption behaviour varies based on biochar properties and mechanisms overcoming this repulsion: For PWBC700, adsorption is hindered by repulsion within pH 6–10. However, when pH 10, despite repulsion, adsorption increases due to the high surface area/pore

structure providing physical sites and the dissolution of ash minerals, releasing cations that bridge the strongly negative HTC²⁻ (Wang et al., 2022), like SWBC700. For NWBC700, when pH is within 5.74–7.3, TCH remains H₃TC while the biochar surface becomes negative. Weak electrostatic attraction or dipole interactions may occur but are insufficient compared to the previous non-electrostatic forces dominant at lower pH, leading to decreased adsorption compared to the 3.3–5.74 range. When pH > 7.3, TCH is mainly H₂TC⁻, and electrostatic repulsion exists. However, strong π - π EDA interactions (between the electron-rich aromatic ring of H₂TC⁻ and the electron-deficient graphitic surface of the biochar) overcome repulsion, coupled with pore-filling, increasing adsorption capacity (Ding et al., 2024). For CaWBC700, MgWBC700, and ZnWBC700, adsorption capacity increases at pH > pHPZC. Precipitates formed at lower pH tend to dissolve under high alkalinity (Zainuddin et al., 2019). The released metal ions (Ca²⁺, Mg²⁺, Zn²⁺) can be complex with HTC²⁻, and these complexes are adsorbed onto the negatively charged biochar surface via metal ion bridging (Jia et al., 2024).

Despite the crucial role of solution pH in the adsorption process, which affects the surface charge of biochar and determines the degree of ionisation of the adsorbate, PWBC700 (48.81–70.83%), SWBC700 (39.45–60.51%), NWBC700 (44.67–60.94%), and MgBC700 (41.89–54.77%) maintained high TCH removal efficiency over a broad pH range (3–10) (see [Supplementary Materials](#)). This finding contradicts *meta*-analysis predictions, which typically indicate maximum adsorption at neutral solution pH, whereas the highest adsorption of TCs by biochar occurred at pH 2, 4, and 10. This discrepancy likely arises because *meta*-analyses, relying on aggregated data from multiple studies, may not fully capture the details of specific experimental conditions. Additionally, while *meta*-analyses emphasise the significant role of solution pH in affecting biochar adsorption performance, the biochar used in the experiments underwent various modifications that could have altered its surface charge characteristics, thereby influencing its adsorption capacity in response to solution pH changes. Additionally, TCs exist in different forms under varying solution pH conditions, and the charge status of the biochar surface also varies with pH. These complex interactions between the adsorbate and the adsorbent may not have been adequately considered in the *meta*-analysis, leading to discrepancies between the predicted and experimental results.

3.3. Kinetic and isotherm experiments for tetracycline hydrochloride adsorption by biochar

In the adsorption experiments, the three biochars with the best adsorption capacities (PWBC700, NWBC700, and SWBC700) and the raw biochar (WBC700) were selected to conduct isothermal adsorption and adsorption kinetics studies. A pseudo-first-order kinetics model was used to simulate the adsorption dynamics of TCH on these biochars. The TCH adsorption capacities of PRBC700, NWBC700, and WBC700 increased rapidly within the first 6 h, gradually reaching equilibrium. In contrast, SWBC700 required 8 h to reach equilibrium (Fig. 5A). Therefore, different modifiers affect biochar differently.

The results of Langmuir, Freundlich, and Sips isotherm models were analysed to elucidate the adsorption mechanisms of TCH on various biochars (see [Supplementary Materials](#), and Fig. 5C-E). The R² values were 0.997 for PWBC700, 0.995 for SWBC700, and 0.988 for NWBC700, indicating a good fit for strong monolayer adsorption. SWBC700 exhibited the highest Q_{max} value of 637.71 mg/g, PWBC700 followed with a Q_{max} of 496.66 mg/g, while NWBC700 showed a Q_{max} of 214.49 mg/g. WBC700 had minimal adsorption capacity with a Q_{max} value of 55.17 mg/g, suggesting limited adsorption potential without modification. Among the four biochars, NWBC700 has the highest k_L value of 0.06 L/g, indicating the fastest adsorption rate. For all samples, the dimensionless separation factor (R_L) values, calculated at an initial TCH concentration of 200 mg/L, were all very close to 1, indicating nearly linear adsorption processes and suggesting spontaneous adsorption.

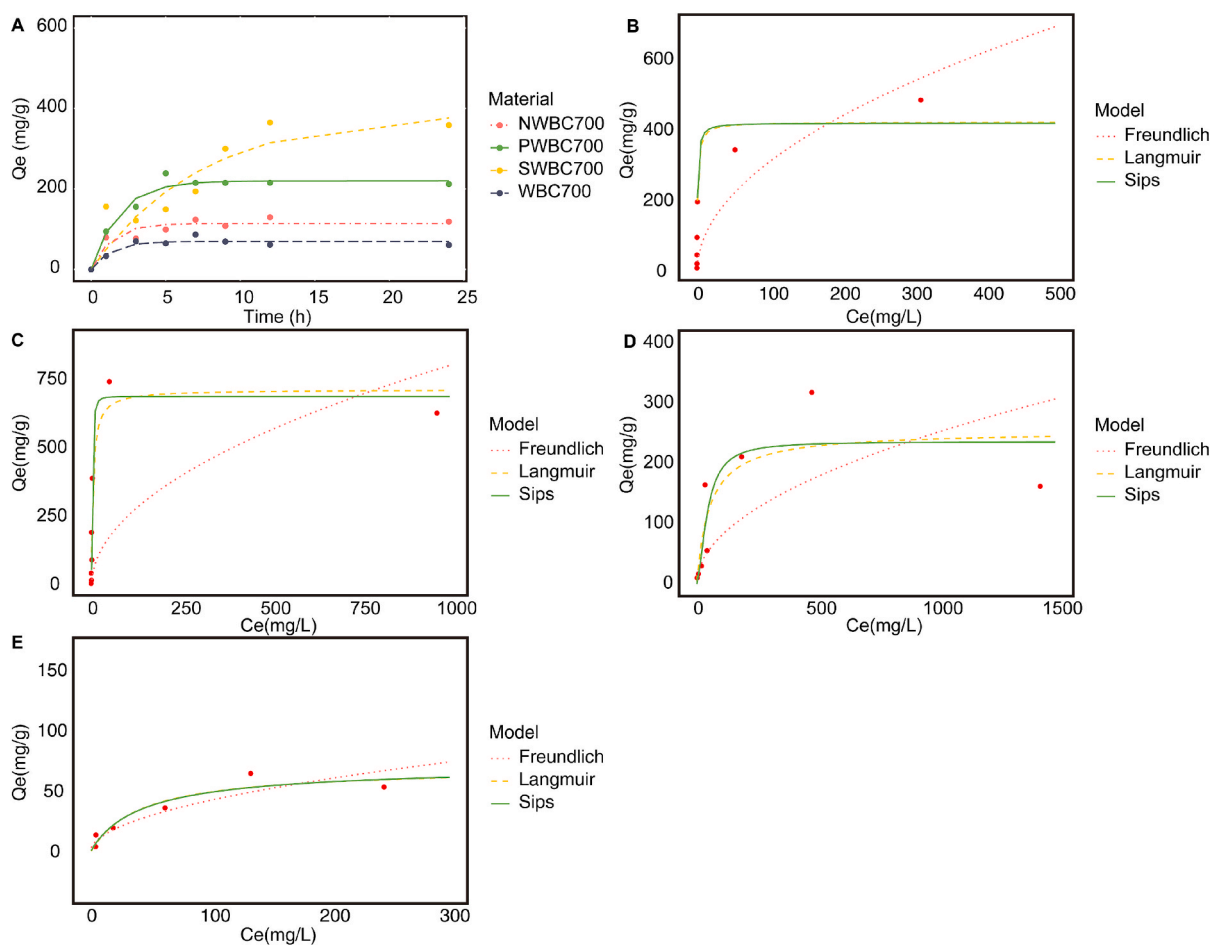


Fig. 5. Adsorption kinetics and isotherm model of modified biochars. Adsorption kinetics of PWBC700, SWBC700, NWBC700, and WBC700 (A) (TCH: 400 mg/L). The linear fitting lines from the Freundlich, Langmuir, and Sips isotherm models for the adsorption of PWBC700 (B), SWBC700 (C), NWBC700 (D), and WBC700 (E).

The $1/n$ values in the Freundlich model are all close to 0.5, indicating favourable adsorption conditions. Among the four biochars, PWBC700 has the highest adsorption rate ($k_f = 23.25$) and the strongest affinity (k_s value = 0.17 L/g) according to the Sips model. While NWBC700 presents complex multilayer adsorption processes ($n = 2$), the data for the other biochars are consistent with heterogeneous surface adsorption. Therefore, PWBC700 is identified as the best-performing sample in terms of adsorption capacity and efficiency.

Considering the previous best-reported value of 543.227 mg/g for TCs (Tan et al., 2020), the materials developed in this research demonstrate significant advantages, as the biochar modified with H_3PO_4 reached a Q_{max} of 540.86 mg/g (Zhang et al., 2023), while H_2SO_4 modification yielded a Q_{max} of 637.71 mg/g, which is substantially higher than the previously reported maximum for TC (92.64 mg/g). These findings underscore the effectiveness of the modifications employed in this study, suggesting that they significantly enhance the TCH adsorption capacity of biochar. Therefore, the model is a highly advantageous guide to biochar preparation.

3.4. Limitations and future perspectives

The limitations of this study include its exclusive focus on plant-based biomass and the use of TCs as the sole model pollutant. Moreover, the *meta-analysis* approach cannot automatically learn patterns or make predictions, which restricts its applicability to a diverse range of antibiotics and the identification of optimal preparation conditions. While TCs serve as the model pollutant in this study, the identified adsorption mechanisms can be selectively applied to other antibiotics

with similar features. Specifically, π - π EDA interactions apply to antibiotics with fused aromatic systems, such as fluoroquinolones through their quinoline rings and sulfonamides via benzene rings (Jia et al., 2024). Cation bridging can enhance adsorption for compounds with metal-chelating groups suitable for salt-modified biochars (Zhang et al., 2023). Meanwhile, antibiotics with pKa values similar to TCs are also applicable in this study. However, the model in this study has limitations to non-analogous antibiotics. For instance, linear β -lactams like penicillin G lack π - π interactions (Lavanya et al., 2013).

To address these limitations, future research could first validate biochar designs on antibiotics grouped by this framework and then attempt to incorporate additional classifications based on antibiotic structure (linear or cyclic), charge type (isoelectric point, anionic, or salt-form), and metal-chelating ability into the analysis. Alternatively, machine learning models could be developed using molecular descriptors to predict adsorption across antibiotic classes as *meta-analysis* can't learn patterns or make predictions, limiting its ability to process diverse antibiotics and find optimal preparation conditions. Such efforts would advance biochar design from compound-specific to class-generalizable solutions for antibiotic pollution control.

4. Conclusion

This study used *meta-analysis* to identify key factors for enhanced TCs adsorption in biochar preparation. Results showed optimal salt/acid-modified biochar performance at temperatures above 400 °C, with straw and wood-based biochars achieving the highest adsorption capacity when pyrolyzed beyond 600 °C. Experiments with pine, poplar,

and crop straws modified by six salts/three acids demonstrated higher biochar adsorption, confirming the *meta*-analysis predictions. Furthermore, biochars prepared under the conditions specified by the models demonstrated higher adsorption capacity than literature reports. However, this study is limited by its exclusive focus on plant-based biomass and a single model pollutant, and the *meta*-analysis approach lacks predictive capability for structurally diverse antibiotics or optimal preparation conditions. Nonetheless, the identified mechanisms are applicable to antibiotics with similar features. Future work should validate these findings across grouped antibiotics based on molecular molecular traits or apply machine learning models to enable broader, structure-informed predictions, advancing biochar design for class-generalizable antibiotic pollution control.

CRedit authorship contribution statement

Na Li: Writing – original draft, Visualization, Methodology, Investigation, Conceptualization. **Xiaochen Zhu:** Visualization, Methodology. **Yahui Miao:** Writing – review & editing, Validation. **Zhenyao Wang:** Writing – review & editing. **Carol Sze Ki Lin:** Writing – review & editing, Supervision, Project administration. **Chong Li:** Writing – review & editing, Visualization, Supervision, Project administration.

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.

Acknowledgements

The authors thank the Natural Science Foundation of Guangdong Province (2025A1515010785) and the Foshan Science and Technology Innovation Project (2320001006040) for funding. They also express gratitude to Dr. Huang Yifan from Wageningen University for his guidance on biochar preparation and adsorption. Professional English language editing support provided by AsiaEdit (asiaedit.com).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.biortech.2025.132917>.

Data availability

Data will be made available on request.

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