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Guiling Gao

Hangzhou Polytechnic University

Haofeng Zhu

Hangzhou Polytechnic University

Ping Wang

Zhejiang Radiation Environment Monitoring Station (Technical Center for Radiation Environment Monitoring)

Lei Hu

Ecological and Environmental Monitoring Center of Zhejiang Province

Xuanyu Zhou

Ecological and Environmental Monitoring Center of Zhejiang Province

Chenghao Wang

Hangzhou Polytechnic University

Zhenyu Chen

Hangzhou Polytechnic University

Zhenya Zhu

dgjj1982@163.com

Hangzhou Polytechnic University

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**Synergistic adsorption of heavy metals by magnetic composite
biochar: Performance and mechanisms in co-contamination systems**

Guiling Gao¹, Haofeng Zhu¹, Ping Wang², Lei Hu³, Xuanyu Zhou³, Chenghao Wang¹,
Zhenyu Chen¹, Zhenya Zhu^{1*}

¹ Hangzhou Polytechnic University, Hang Zhou, 310018, P.R. China

² Zhejiang Radiation Environment Monitoring Station (Technical Center for Radiation
Environment Monitoring, Ministry of Ecology and Environment), Hangzhou, 310012,
P. R. China.

³ Ecological and Environmental Monitoring Center of Zhejiang Province, Hangzhou
310012, P. R. China

*Corresponding author: Zhenya Zhu

E-Mail: dgjj1982@163.com

Address: Xueyuan Street No. 68, College of Chemical and Biological Engineering,
Hangzhou Polytechnic University, Qiantang District, 310018 Hangzhou, P. R. China

Abstract

Heavy metals and antibiotics often coexist in aquatic environments, posing complex challenges for water purification. In this study, magnetic composite biochars (M-AMK) was successfully fabricated via grinding-assisted pyrolysis and hydrothermal methods. Characterization results revealed that M-AMK possessed a well-developed porous structure with an average pore diameter of 23.68 nm, uniformly dispersed Fe₃O₄ nanoparticles, and abundant surface functional groups, including –OH, C=C, C–O, and Fe–O. In the Pb-TC binary system, M-AMK exhibited strong preferential adsorption for Pb(II), with removal efficiency reaching 99.8% and remaining high even with coexisting TC, owing to stronger interactions with oxygen-containing groups and Fe⁰ sites. The synergistic mechanisms included ion exchange, surface complexation, precipitation, and π – π interactions, with the formation of TC–Pb complexes further promoting stability and reducing metal leaching. The magnetic composites also demonstrated excellent reusability and structural stability over multiple cycles. This work provides a promising strategy for designing multifunctional adsorbents capable of efficient and selective removal of combined pollutants in complex water matrices.

Keywords: Magnetic composite biochar; Pb(II); Tetracycline; Binary co-contamination; Adsorption mechanism

1. Introduction

With the rapid development of industrial and agricultural activities, water

43 pollution has become increasingly severe(Robert E Dorsey *et al.* 2010;Cüce, H. *et*
44 *al.*2022). In particular, the co-contamination caused by the simultaneous discharge of
45 heavy metals and antibiotics has emerged as a global environmental challenge(hu
46 Lizhong *et al.* 2000;Topaldemir, H.. *et al.* 2023). Heavy metals (e.g., lead, cadmium,
47 copper) possess bioaccumulative and toxic properties, while antibiotics (e.g.,
48 tetracycline, sulfonamides) are prone to inducing microbial resistance(Dong, J. *et al.*
49 2022; KYu *et al.* 2017;Deng, H. *et al.* 2022). The coexistence of these pollutants not
50 only complicates water treatment but may also lead to more intricate ecological and
51 health risks through synergistic or antagonistic interactions(Komijani, M.*et al.* 2021).
52 Conventional treatment technologies designed for single pollutants are often
53 inadequate in effectively addressing such multi-component and interactive
54 contamination systems. Among various water treatment technologies, adsorption has
55 attracted considerable attention due to its high efficiency, operational simplicity, and
56 relatively low cost. Biochar, as a green and porous carbonaceous adsorbent, has
57 demonstrated promising potential for the removal of heavy metals and organic
58 pollutants(Suo F. *et al.* 2021). However, traditional biochar often suffers from limited
59 adsorption capacity, low selectivity, and difficult recovery when dealing with
60 composite pollution(Shakeel Ahmad *et al.* 2023; Kong, Q.*et al.* 2021). In recent years,
61 the construction of composite biochar materials by incorporating mineral components
62 (e.g., kaolin, montmorillonite) and magnetic phases (e.g., Fe₃O₄) has become an
63 important strategy to enhance adsorption performance and material

64 functionality(Cheng Dongle *et al.* 2020; Khorshidi Parisa *et al.* 2020; Liuwei Wang *et*
65 *al.* 2026). Such composites can integrate multiple adsorption mechanisms (e.g., ion
66 exchange, surface complexation, π - π interactions, electrostatic attraction(Wang L *et al.*
67 2022 C)) and enable rapid solid-liquid separation via magnetism, showing favorable
68 application prospects(Khorshidi Parisa *et al.* 2020;Li Xuhui *et al.* 2020).

69 Nevertheless, systematic understanding of the synergistic adsorption behavior
70 and mechanisms of magnetic composite biochar in heavy metal-antibiotic
71 co-contamination systems remains insufficient(Rai N *et al.* 2025). In particular, the
72 competitive adsorption mechanisms among pollutants, the role of surface active sites,
73 and the influence of microstructure on adsorption performance are still not well
74 elucidated. Therefore, developing multifunctional composite materials with high
75 adsorption efficiency and good magnetic recoverability, and thoroughly investigating
76 their removal performance and mechanisms in composite pollution systems, is of
77 significant scientific importance for advancing practical wastewater treatment
78 technologies.

79 Previous study successfully synthesized a composite biochar material derived
80 from river/lake sediment(Bai Xue *et al.* 2021), microalgae(Xu Ling 2010), and kaolin
81 through a green grinding-co-pyrolysis-in situ oxidation method (designated as AMK).
82 In this study, Fe_3O_4 was successfully loaded onto the composite biochar through an
83 innovative two-step method of “grinding-in situ oxidation,” yielding a magnetic
84 multifunctional composite biochar material (designated as M-AMK)(Chen, D. *et al.*

2017;Wang, S. *et al.* 2019). The adsorption performance of the material for typical heavy metals (e.g., Pb(II)) in both single and binary systems will be systematically evaluated. Combined with multiple characterization techniques and adsorption modeling, the microstructural features and multi-mechanistic adsorption pathways will be elucidated, aiming to provide new materials and theoretical foundations for the control of complex water pollution.

2. Materials and methods

2.1 Materials

Fe₃O₄ was purchased from Sinopharm Chemical reagent Company(Shanghai, China). Tetracycline (TC) was obtained from Macklin. Algae sludge was salvaged from DianChi Lake. All agents were of analytical-reagent grade. Deionized Millipore Milli-Q (18 MΩ/cm) water was used throughout the experiment.

2.2 Preparation of M-AMK

A predetermined mass ratio (5:1) of the composite biochar (AMK) to Fe₃O₄ was placed in a mortar and ground solvent-free for 30 minutes to ensure uniform dispersion of Fe₃O₄ on the biochar surface and within its pores. The resulting mixture was then transferred to a muffle furnace and maintained at 200°C for 1 hour to enhance the binding stability between the magnetic component and the biochar matrix, thereby preventing detachment of magnetic particles during subsequent use. After cooling to room temperature, the sample was treated with 50 mL of a 1:10 HCl solution for 30 minutes to remove surface-free Fe³⁺/Fe²⁺ ions, rinsed with distilled water until neutral, dried at 80°C, ground, and sieved through a 100-mesh sieve to

obtain the magnetic composite biochar material, labeled as M-AMK.

2.3 Characterization methods

The phase composition of the materials was analyzed by X-ray diffraction (XRD, Bruker D8 Advance). The surface morphology and elemental distribution of the composite biochar were observed using transmission electron microscopy (TEM). Magnetic properties were measured with a vibrating sample magnetometer (VSM, Lakeshore 7404). The surface areas and nitrogen adsorption-desorption isotherms of the samples were determined by a BELSORP-Mini II analyzer and calculated by Brunauer-Emmett-Teller (BET) method. Surface functional groups of the composite biochar were characterized by Fourier-transform infrared spectroscopy (FT-IR).

2.4 Competitive adsorption affected pH

(1) Competitive adsorption of Pb(II) as affected by TC and pH

To evaluate the competitive adsorption of Pb(II) and TC on the M-AMK composite, batch adsorption experiments were conducted. All tests were conducted in 250 mL conical flasks by dosing 0.5 g/L of M-AMK composite in the solution with a fixed initial Pb(II) concentration of 50 mg/L and changing TC concentrations from 0 to 30 mg/L. The effect of pH on adsorption as affected by TC was also conducted. The suspension pH was adjusted to different values between 2 and 7 using dilute HCl or NaOH. Adsorption was conducted by shaking at 170 rpm for 180 min at 30 °C. After every 5 min, 2 mL sample was pipetted and used for metal detection through atomic absorption spectrometer on flame mode. The experiments were performed in

duplicate.

Adsorption experiments performed with these solutions at three different temperatures (15 °C, 25 °C, 35 °C). The removal efficiency (R%) is calculated in Eq.

(1):

$$R \% = \frac{c_0 - c_t}{c_0} \times 100 \quad (1)$$

The amount of adsorbed is calculated in Eq. (2):

$$q_t = \frac{(c_0 - c_t) \times V}{m} \quad (2)$$

where c_0 and c_t are the initial and equilibrium concentration (mg/L). V is the volume of suspension solution (L), m is the mass of adsorbent dosed (g).

It is more common practice for adsorption kinetic models to be used to fit adsorption experimental data. The linear equations of pseudo-firstorder dynamics (3) and pseudo-second-order dynamics (4) are as follow:

$$\frac{1}{Q_t} = \frac{K_1}{tQ_e} + \frac{1}{Q_e} \quad (3)$$

$$\frac{t}{Q_t} = \frac{1}{K_2 Q_e^2} + \frac{t}{Q_e} \quad (4)$$

Where Q_t is unit adsorption capacity at time t , mg/g; K_1 is pseudo first-order kinetic model constant, min^{-1} ; K_2 is pseudo-second-order kinetic model constant, $\text{gmg}^{-1}\text{min}^{-1}$; Q_e is unit adsorption capacity at adsorption equilibrium, mg/g.

2.5 Adsorption mechanism analysis

The adsorption of heavy metals by biochar involves multiple mechanisms, typically including: (1) ion exchange with cations; (2) precipitation with inorganic substances; (3) complexation with oxygen-containing functional groups; and (4)

coordination with π electrons. By acid washing and mineral leaching, most minerals are removed from the biochar material with the surface functional groups unchanged. The decrease in adsorption capacity before and after acid washing can be considered as the adsorption capacity of minerals action (Q_{cm}). Exchangeable cations (K^+ , Ca^{2+} , Na^+ , and Mg^{2+}) in the biochar materials participate in ion exchange reactions. Therefore, the quantity of cation exchange (Q_{cme}) can be calculated based on the change of these cations before and after adsorption. The adsorption of minerals is a combined effect of cation exchange and mineral precipitation. Therefore, the quantity of mineral precipitation (Q_{cmp}) can be calculated as the difference between the adsorption of minerals (Q_{cm}) and the quantity of cation exchange (Q_{cme}) (Zhong Xiaoxiao *et al.* 2019), according to Eqs. (1) - (2) :

$$Q_{cm} = Q - Q_a \times Y \quad (1)$$

$$Q_{cmp} = Q_{cm} - Q_{cme} \quad (2)$$

Where Q_{cm} , Q_{cmp} , and Q_{cme} are the adsorption contributions of mineral action, precipitation, and cation exchange, respectively ($mg \cdot g^{-1}$); Q is the adsorption capacity pre-acid-washing, and Q_a is the adsorption capacity of the post-acid-washing ($mg \cdot g^{-1}$); Y is the yield of the acid-washed biochar (%).

During the adsorption process, the complexation of Pb(II) with oxygen-containing functional groups release H^+ , causing a decrease in the pH value (Xu Piao *et al.* 2012). Therefore, the contribution of functional group complexation (Q_{co}) can be calculated based on the decrease in pH value of the solution after adsorption. The contribution of π -electron coordination ($Q_{c\pi}$) can be obtained by subtracting the adsorption capacity of acid-washed biochar from its contribution value of oxygen-containing functional groups, according to Eqs. (3) - (4) :

$$Q_{cm} = Q - Q_a \times Y \quad (3)$$

$$Q_{cmp} = Q_{cm} - Q_{cme} \quad (4)$$

Where Q_{co} is the adsorption contribution of functional group complexation, while $Q_{c\pi}$ is the adsorption contribution of π - electron coordination.

2.6 Sequential lead extractions

Sequential extraction were conducted on the adsorbed biochar material to investigate the stability of lead on the surface of biochar after adsorption. The lead adsorbed can be divided into three forms: exchangeable, acid-soluble, and non-bioavailable. The exchangeable form has high mobility and poses a higher risk; the acid-soluble form has lower mobility; whereas the non-bioavailable form exhibits strong stability with low risk(Wang Tongtong *et al.* 2020).

The extraction experiment was conducted as follows: (1) weigh 0.1 g of the adsorbed sample into a 50 mL polyethylene tube, add 8 mL of 0.5 mol·L⁻¹ MgCl₂ (adjusted to pH 7.0 using NaOH or HCl), and shake for 20 minutes for extraction. (2) Similarly, weigh 0.1 g of the adsorbed sample into another polyethylene tube, add 8 mL of 1 mol·L⁻¹ NaOAc (adjusted to pH 5.0 using HOAc), and shake for 5 hours for extraction. (3) Collect the supernatant after shaking, filter through 0.22 μm membrane, and measure the concentration of Pb(II) . All experiments are conducted in triplicate.

The content of exchangeable lead can be obtained through step (1). The sum of exchangeable and acid-soluble lead contents can be obtained through step (2). The non-bioavailable lead content can be obtained by subtracting the sum of exchangeable and acid-soluble lead from the total lead adsorbed by the sample.

2.7 Recyclability analyses

To assess the stability and reusability of the catalysts, the identical binary system were conducted across 5 consecutive reactions. After each cycle, the catalyst was separated using an external magnet, washed, vacuum dried, and subsequently employed for the subsequent sequential experimentation.

3. Results and discussion

3.1 Structure and composition analysis

The surface morphologies of the biochars were examined using scanning

electron microscopy (SEM).

As shown in Fig.1 (1), The sediment-derived biochar (1a) exhibited a porous and loose structure, while the algal-derived biochar (1b) showed an aggregated, floc-like porous morphology. The kaolin-derived biochar (1c) displayed a distinct lamellar architecture. In contrast, the AMK composite biochar (1d) possessed a compact yet non-aggregated texture with the formation of amorphous clustered pores. After magnetic modification, the M-AMK composite presented a well-developed porous network and a relatively smooth surface, which is likely attributed to the intercalation and surface modification by iron species. The in-situ attachment of numerous fine and uniformly dispersed particles further indicates that the composite effectively suppresses the agglomeration of metal oxides, resulting in a highly porous microstructure(Ibrahim MM *et al.* 2023b).

Energy-dispersive X-ray spectroscopy (EDS) analysis (Fig.1 (2)) confirmed that M-AMK is primarily composed of C, Fe, Si, Na, and Al. The dominant C signal reflects the carbon-rich nature of the biochar matrix. The distinct Fe-K α and Fe-K β peaks verify the successful loading of the Fe₃O₄ magnetic phase. The presence of Si and Al originates from the kaolin raw material, while Na is derived from mineral components in the sediment. These results demonstrate that all starting materials contributed to the composite formation, aligning with the “treating-waste-with-waste” design concept.

Elemental mapping images (Fig.1 (3)) further revealed the spatial distribution of

key elements within M-AMK. Carbon was uniformly and extensively distributed, indicating a continuous biochar matrix. Iron was concentrated within the material domain without obvious agglomeration, confirming Fe-containing particles were relatively homogeneously distributed over the observed region. The distribution regions of Si, Na, and Al closely overlapped with that of C, suggesting thorough mixing of the raw components during pyrolysis and grinding, with no noticeable phase separation. This homogeneity contributes to the structural stability and performance consistency of the composite.

3.2 Surface area and pore distribution

The pore characteristics of the M-AMK composite were evaluated using N₂ adsorption-desorption analysis (Fig.1 (4-5)). The isotherm exhibited a type-IV curve with an H3-type hysteresis loop across a wide relative pressure range, indicating the presence of slit-shaped pores (Fig.1 (4)). The calculated average pore diameter was 23.68 nm, confirming the mesoporous nature of the material. Pore size distribution analysis further revealed a hybrid pore system containing both micropores and mesopores (2 – 50 nm) (Fig.1 (5)), which is beneficial for mass transfer and pollutant adsorption.

The surface functional groups of M-AMK were characterized by FT-IR spectroscopy (Fig.1 (6)). The spectrum shows characteristic bands corresponding to –OH/N–H (3424.89 cm⁻¹), which participate in hydrogen bonding with polar organics and complexation with heavy metal ions such as Pb²⁺ and Cd²⁺ (Hameed Rashida *et al.*

2019). The peak at 1625.81 cm^{-1} is attributed to C=C stretching(Ye Xiong *et al.* 2021), enabling π - π interactions with aromatic pollutants. The band at 1036.93 cm^{-1} corresponds to C-O/Si-O vibrations, contributing to electrostatic attraction and chemical bonding with cationic metals(Liu Meng *et al.* 2022). Additionally, the presence of Fe-O bonds (near 465 cm^{-1}) confirms the successful loading of Fe_3O_4 (Xu Piao *et al.* 2012; Saravanan A *et al.* 2023), which not only provides magnetic separability but also facilitates ion exchange and radical-mediated degradation of organic pollutants, thereby combining adsorption with catalytic function(Weiquan Cai *et al.* 2019).

3.3 Magnetic Property Analysis

The magnetic properties of the composite were evaluated through vibrating sample magnetometry at room temperature. As shown in Fig.1 (7), the magnetization of M-AMK increased with the applied magnetic field until reaching saturation. The measured saturation magnetization was 37.36 emu/g , confirming the successful incorporation of a magnetic phase.

The recoverability of M-AMK under an external magnetic field was also examined. Fig.1 (7) also illustrates that after 3 minutes of magnetic exposure, the dispersed particles rapidly accumulated toward the magnet, demonstrating efficient magnetic separation. These results indicate that the composite possesses sufficient magnetic responsiveness for practical recovery and reuse in aqueous systems.

3.4 Adsorption in binary system

(1) Competitive adsorption

The competitive adsorption between Pb(II) and TC was evaluated by varying the concentration of one pollutant while keeping the other fixed at 50 mg/L. As shown in Fig. 2(a), M-AMK exhibited significantly higher adsorption capacity for Pb(II) compared to the non-magnetic AMK. The presence of TC had a minimal effect on Pb(II) removal by M-AMK, with removal efficiency remaining consistently high (79 – 99%). This indicates a stronger affinity of Pb(II) towards the functional groups and Fe⁰ sites on M-AMK. The highest removal was observed at a TC concentration of 10 mg/L, likely due to the formation of TC – Pb complexes which reduce free Pb(II) in solution. Further increase in TC concentration led to competitive site occupation and steric hindrance, slightly suppressing Pb(II) adsorption.

(2) Effects of pH

The influence of initial pH on Pb(II) adsorption in the binary system is presented in Fig. 1(b).

At low pH (<3), the positively charged adsorbent surface repelled Pb²⁺ cations, resulting in lower removal. As pH increased (3 – 5), the surface became negatively charged, enhancing adsorption through electrostatic attraction and complexation with surface functional groups(Shakeel Ahmad *et al.* 2020). The presence of TC further improved Pb(II) removal in this range via a bridging effect. At pH > 5, the predominant process shifted to Pb(OH)₂ precipitation in the single system. In contrast, in the binary system, the formation of Pb precipitates was partially hindered

by complexation with TC, thereby causing a progressive decline in adsorption performance(Liang Jie *et al.* 2020).

3.5 Adsorption mechanism analysis

(1) Adsorption kinetic study and Equilibrium adsorption isotherms can be referenced in the supplementary information.

(2) Pb(II) adsorption mechanisms in binary system

① *Surface morphology before and after adsorption*

SEM and EDS mapping (Fig. 3 (1)) revealed the aggregation of surface particles on M-AMK after Pb(II) adsorption, suggesting the formation of surface complexes. The uniform distribution of Pb in the elemental map confirmed successful and homogeneous adsorption across the material's active sites.

XRD patterns (Fig. 3 (2)) showed that the crystal structure of M-AMK remained stable after adsorption, with a slight increase in interlayer spacing (from 1.85 nm to 1.93 nm), indicating partial intercalation of Pb. The appearance of PbSO₄ diffraction peaks suggests precipitation and surface complexation(Guo Sheng *et al.* 2020). FT-IR spectra (Fig. 3 (3)) displayed a significant decrease in –OH and carboxyl group peaks after adsorption, indicating the involvement of π electrons in the adsorption process. Besides, the emergence of a Pb–O vibration peak at 630 cm⁻¹, confirming the involvement of oxygen-containing groups in Pb(II) binding via complexation and ion exchange(Elmolla E S *et al.* 2010).

DFT modeling (Fig. 3 (4)) indicated strong Pb–O coordination, consistent with

experimental findings. Density of states analysis (Fig. 3(5-7)) showed that adsorption did not alter the overall electronic structure of M-AMK, with Fe and O orbitals dominating the frontier region, supporting their key role in chelating Pb(II)(Saravanan A *et al.* 2023). Zeta potential measurements (Fig. 3 (8)) indicated an increase in the point of zero charge after Pb adsorption, consistent with consumption of surface anions and hydroxyl groups.

② Adsorption mechanism analysis in binary system

Compared to the initial pH value 5.2, the pH of the binary system decreased to 4.4 after adsorption, which was less than the decrease in the single system (to pH 3.6). Tetracycline (TC) is an amphoteric molecule whose speciation and surface charge vary with solution pH, thereby influencing Pb adsorption behavior. Under highly acidic conditions ($\text{pH} < 3$), the abundance of hydronium ions (H_3O^+) protonates the electron-rich π -systems on the adsorbent surface, resulting in a net positive surface charge(Zhou Jiahui *et al.* 2022). At this pH, electrostatic repulsion dominates, and no significant TC–Pb complexes are formed; instead, both species compete for adsorption sites, leading to reduced removal efficiency(Ma, J. *et al.* 2020).

In the moderately acidic range ($3 < \text{pH} < 5$), TC exists predominantly in a cationic form. Functional groups such as phenolic, amino, hydroxyl, and ketone moieties can undergo specific interactions (e.g., hydrogen bonding, complexation) with the composite material. Within this pH window, the affinity of TC–Pb complexes for the adsorbent surface is stronger than that of free TC molecules, favoring

adsorption.

At higher pH ($\text{pH} > 5$), TC adopts a zwitterionic or anionic form. Electrostatic repulsion between the negatively charged TC species and the likewise negatively charged adsorbent surface reduces adsorption efficiency. In the binary system, Pb(II) begins to hydrolyze, forming $\text{Pb}(\text{OH})^+$ and $\text{Pb}(\text{OH})_2$ species under alkaline conditions(Elmolla E S *et al.* 2010). These hydrolyzed Pb species can electrostatically attract anionic TC, while TC and its complexes can partially inhibit the precipitation of Pb-containing solids.

Quantitative analysis (Fig. 4a) showed that as TC concentration increased, the contribution of complexation rose (20.07% to 52.94%), while those of precipitation and ion exchange declined. Sequential extraction (Fig. 4b) indicated that most adsorbed Pb was stabilized in acid-soluble and non-bioavailable forms, with stability enhancing at higher TC concentrations due to strong TC–Pb complexation, thereby reducing leaching risk(Liang Jie *et al.* 2017).

③ Recycling and stability experiments

After five cycles, Pb(II) removal efficiency declined from 99.8% to 78.8% and from 95.26% to 89.18% in the binary system and in the single system, respectively (Fig. 5 (1-2)). In the binary system, one heavy metal - antibiotics complex may chelate with multiple metal ions, resulting in a higher removal efficiency. Besides, the Fe^0 on the surface of the M-AMK reacted with molecular oxygen, producing more reactive oxygen species for TC degradation and inturn prompting adsorption

process(Chen Yong *et al.* 2011). SEM images (Fig. 5 (3)) showed increased aggregation but maintained structural integrity. Atomic Absorption Spectroscopy (AAS) technology was applied to investigate Iron leaching during the Pb(II) adsorption (Fig. 5 (4)). Iron leaching in binary system (0.25 mg/L) was lower in the single system (0.36 mg/L), indicating that TC–Fe interactions further stabilized the composite.

Conclusion

In this study, magnetic composite biochars (M-AMK) was successfully fabricated via a grinding-assisted pyrolysis-hydrothermal method for the simultaneous removal of Pb(II) in binary pollution systems. The composites exhibited a well-developed pore structure, abundant surface functional groups, and uniformly dispersed Fe₃O₄ nanoparticles, which endowed them with excellent adsorption capacity and magnetic separability.

The results demonstrated that Pb(II) was preferentially adsorbed with high efficiency (up to 99.8%) even in the presence of TC, indicating a stronger affinity between Pb(II) and the composite surface. pH played a critical role in regulating adsorption performance by affecting the speciation of TC and the surface charge of the composite.

Mechanistic studies revealed that the removal process involved multiple synergistic pathways, including ion exchange, surface complexation with

oxygen-containing groups, precipitation, π - π interactions, and the formation of stable TC–Pb complexes. These complexes not only promoted adsorption but also reduced the risk of Pb leaching, improving the environmental stability of the adsorbed contaminants.

This work provides a feasible strategy for designing multifunctional magnetic biochar composites capable of efficiently treating co-existing heavy metal and antibiotic pollutants. The insights gained into the synergistic adsorption mechanisms offer a scientific basis for the application of such materials in complex wastewater environments. Future work will focus on investigating the degradation performance and removal mechanisms of antibiotics by composite biochar materials in contaminated systems.

CRedit authorship contribution statement

Guiling Gao: Investigation, Formal analysis, Writing-original draft. **Haofeng Zhu:** Data curation, Validation. **Ping Wang:** Sample collection, Data curation, Validation. **Lei Hu & Xuanyu Zhou:** Conceptualization, Formal analysis. **Chenghao Wang & Zhenyu Chen:** Conceptualization, Funding acquisition, Writing-review & editing. **Zhenya Zhu:** Methodology, Supervision, Funding acquisition.

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395 Composite Biochar Materials and Study on Photocatalytic Degradation of Algal
396 Toxins” (Grant No. ky202533).

397

398 **Declaration of competing interest**

399 The authors declare no competing financial interest.

400

401 **Data Availability**

402 The dataset generated and analyzed during the current study are available from the
403 corresponding authors on reasonable request.

404

405

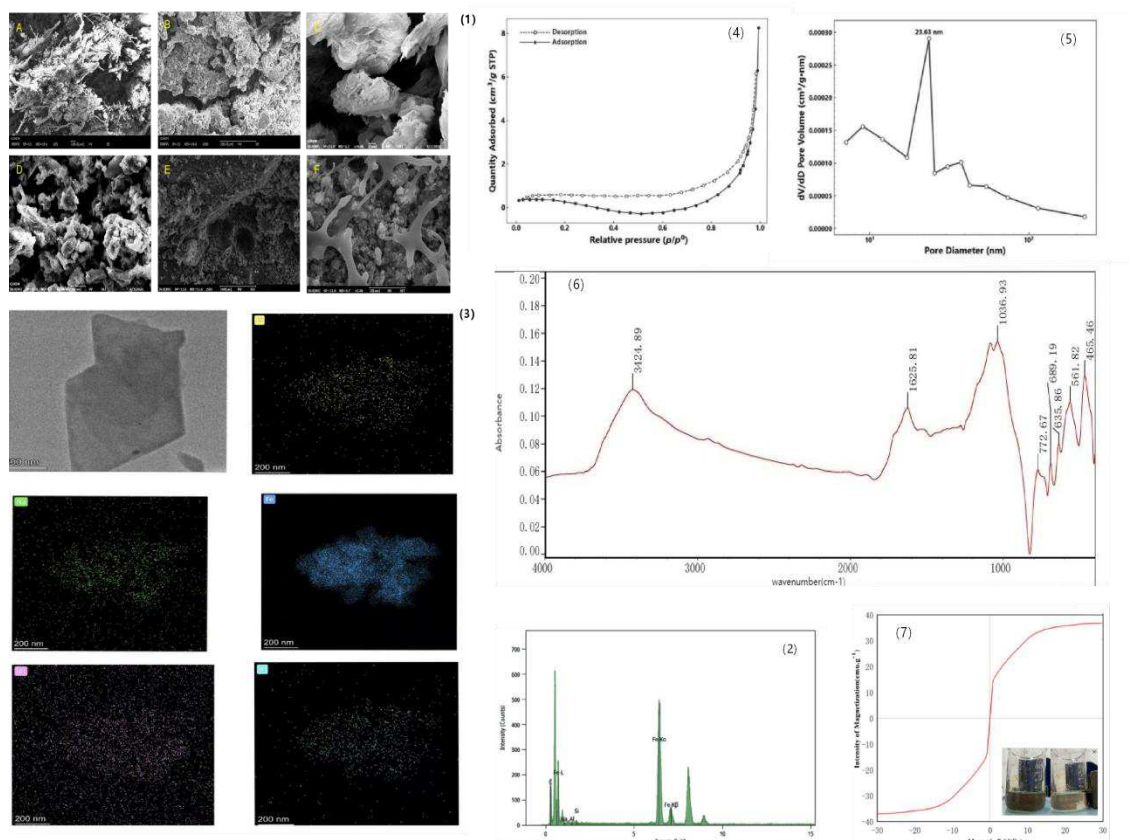


Fig. 1 (1) SEM images of (a) sediment biochar , (b) algal biochar, (c) kaolin biochar, (d)AMK composite biochar,(e)(f) M-AMK composite biochar; (2) EDS spectrum; (3)Element mapping images; (4)(5) specific surface area and pore distribution; (6) FT-IR spectra of M-AMK;(7) Magnetic hysteresis loop and the response of the dispersion to magnetic field

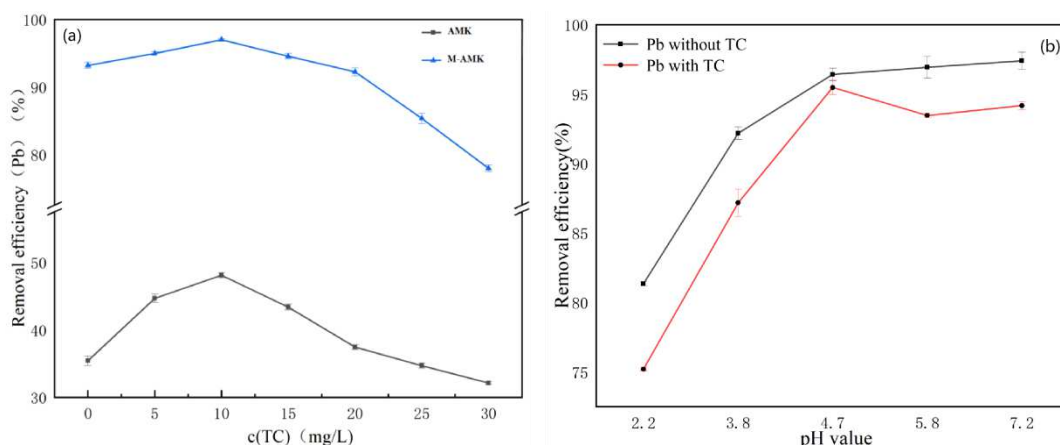


Fig. 2 Relationship between Pb(II) adsorption efficiency and TC concentration(a) and relationship between pH values and Pb(II) adsorption efficiency(b)
($c_0(\text{Pb})=50 \text{ mg/L}$, $c_0(\text{TC})=10 \text{ mg/L}$, 30°C , dosage= 0.5 g/L)

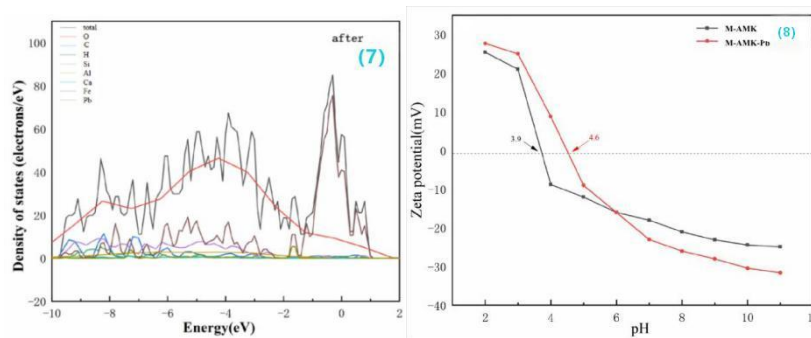
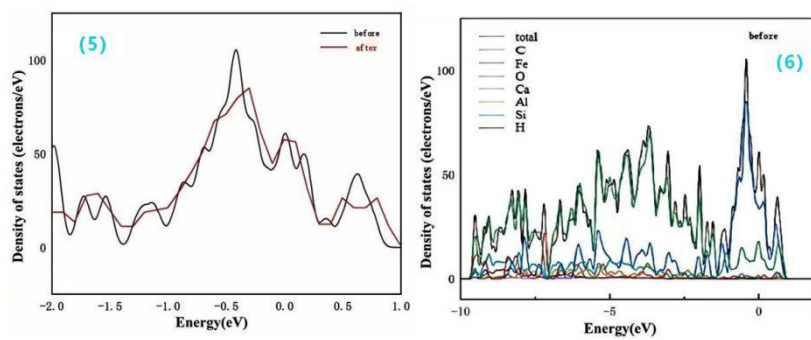
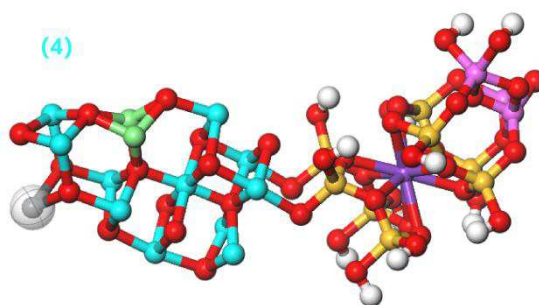
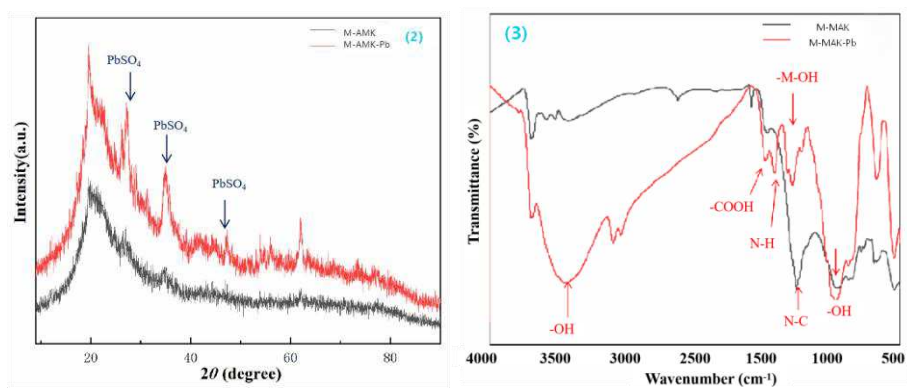
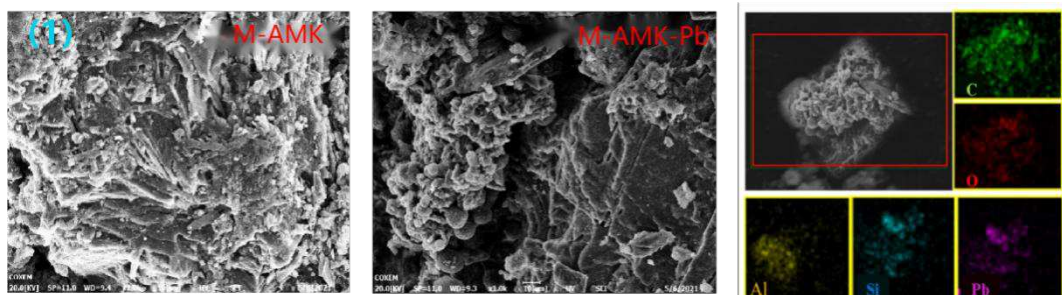


Fig. 3 (1) SEM images of M-AMK and M-AMK-Pb and the scanning transmission electron microscope-high angle annular dark field image of M-ZnO-AMK after Pb(II) adsorption along with EDS elemental mapping of C、O、Al、Si、Pb; (2) XRD and (3) FT-IR spectra of M-AMK and M-AMK-Pb; (4) The molecular structure of M-AMK-Pb; (5) Total density states、(6-7) partial atomic density of states and (8)Zeta potential before and after adsorption

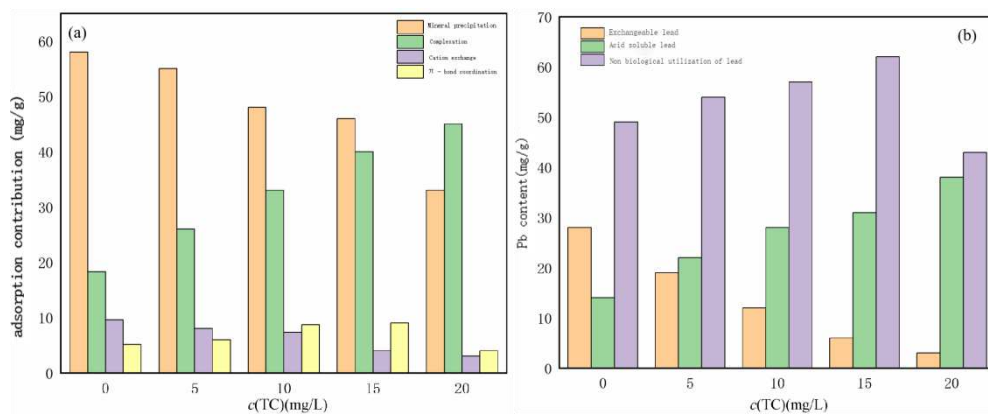


Fig. 4 The contribution rate of each mechanism and proportion of lead in binary system

($V=100$ mL, 30°C , dosage=50 mg, $c_0(\text{Pb(II)})=50$ mg/L, pH 5.0)

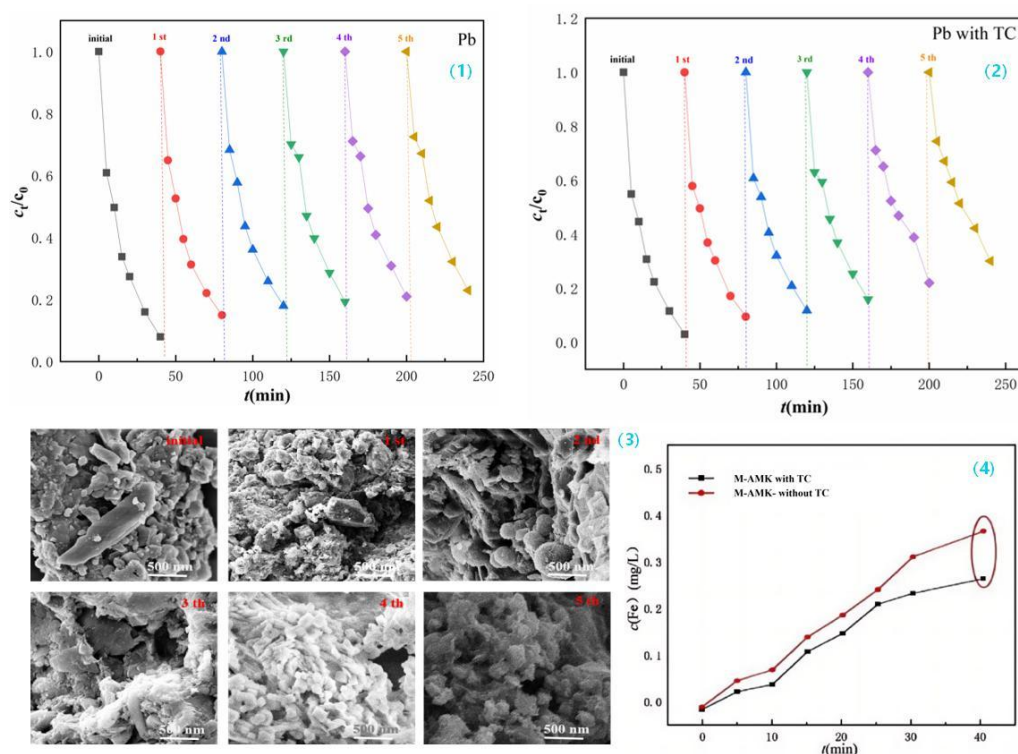


Fig. 5 The relationship between the number of experimental repetitions and Pb(II) concentration over time in single (1) and in binary (2) system; (3) SEM images of M-AMK in different repetitive experiments; (4) Iron leaching concentration in single and in binary system ($c_0(\text{Pb(II)})=50 \text{ mg/L}$; $\text{pH } 5.0$, $30 \text{ }^\circ\text{C}$, $c_0(\text{TC})=10 \text{ mg/L}$)

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