



Inhibiting release of phenanthrene from rice-crab coculture sediments to overlying water with rice stalk biochar: Performance and mechanisms

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Research significance

- Bioturbation is an important factor in the secondary release of contaminants from the sediments. Clarifying the mechanism of crab bioturbation on the release of PAHs from paddy field sediments is an important basis for correctly assessing the ecological risk of rice-crab co-cropping production model.



Rice and Crab Cultivation Income Increases by About Two Times

Comprehensively implement the requirements of the Fourteenth Five-Year Plan and promote the high-quality development of the rice-fishing industry.



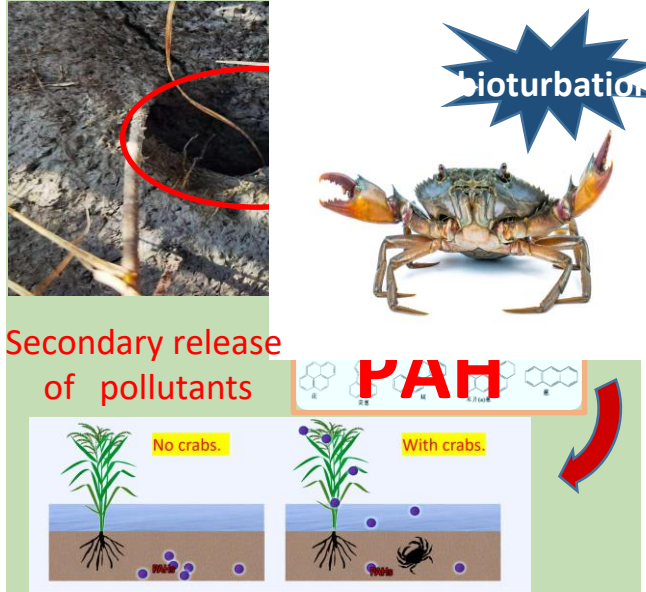
Rice and crab farming became one of the priorities in the structural adjustment of the agricultural industry due to its significant income-generating benefits.



Variety Selection Irrigation pattern fertilizer ratio

Farming management Tillage measures Reduction of pesticides

Currently, it only focused on enhancing the experimentation and research on the rice-crab co-cropping eco-agricultural system, while ignoring the potential risks posed by bioturbation.



bioturbation

Secondary release of pollutants

PAH


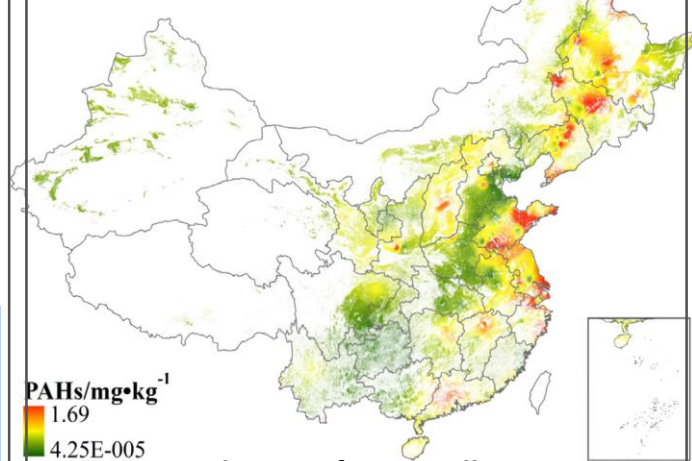
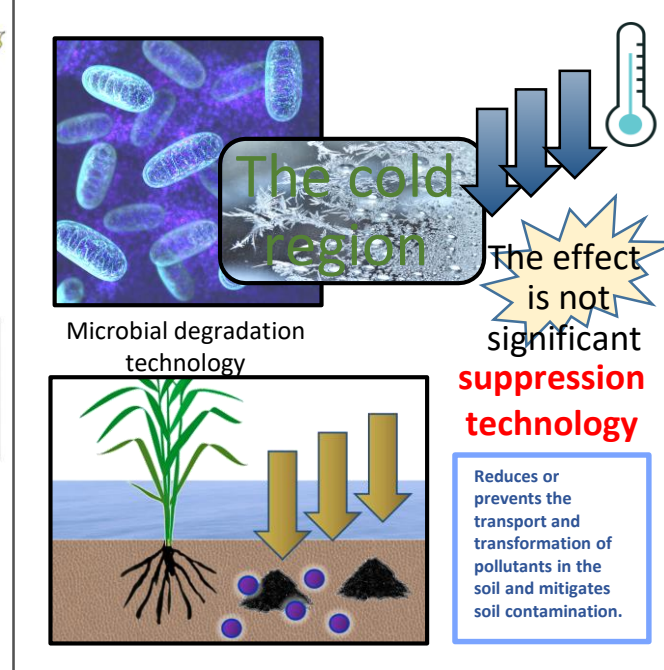
No crabs. With crabs.

Bioturbation refers to various activities of benthic animals, such as feeding, burrowing, defecation, and migration. Bioturbation was prone to secondary releases of contaminants from the sediments.



Research background

- Inhibition the release of PAHs from the sediment of rice-crab co-cultivation paddy fields in the Northeast alpine region is an important way to reduce the ecological risk, and guarantee the green agriculture and food safety.

<p>As the "ballast" of China's food security, the total grain output of Heilongjiang Province in 2021 exceeded 150 billion pounds, accounting for 11.5% of the country's total grain output.</p>		 <p>Distribution of PAHs pollution in China by region</p> <p>PAHs were found as high as 355 $\mu\text{g}/\text{kg}$ in soil and sediment of Heilongjiang farmland, which was higher than organic pollutants such as pesticides and reached a moderate pollution level.</p>	 <p>The cold region</p> <p>The effect is not significant</p> <p>Microbial degradation technology</p> <p>suppression technology</p> <p>Reduces or prevents the transport and transformation of pollutants in the soil and mitigates soil contamination.</p>
<p>Heilongjiang Province was an important grain-producing area in China, with 60 million mu of rice planted annually, ranking first in the country." Rice-crab co-cropping" has become a key recommended production model.</p>	<p>PAHs were common pollutants in agricultural fields in Heilongjiang. Secondary release of PAHs under crab bioturbation entered the food chain via the water-soil-plant-animal pathway, jeopardizing human health and food security.</p>	<p>The climatic characteristics of the cold region make microbial degradation of PAHs less effective. Therefore, the inhibitory technology to control PAHs release should be the first choice for remediation.</p>	



Technical Optimization

- Research on control technology of PAHs release from bottom mud pollutants

Technologies for controlling the migration and release of bottom mud pollution mainly include: ① pollutant removal technologies (e.g. microbial remediation) ② pollutant inhibition technologies (in-situ and ex-situ inhibition technologies).

① Pollutant removal technology

Pollutants Removal technologies (e.g., microbial, etc.)

The Cold Region

insignificant

Biodegradation removal techniques were ineffective in the cold northeast region for low temperature.

Inhibition technology has become hotspot for controlling the release of pollutants from bottom mud

② Pollutant Suppression Technology

Ectopic suppression technology is costly, and needs large space requirement.

In-situ suppression technology has better feasibility and applicability.

✓ In situ suppression technology

× Anisotropic suppression technology

In-situ control technology is subdivided into: adsorbent in-situ modification, In situ active cover, etc.

Active cover technology is affected by benthic disturbance.

In-situ improvement technology has more advantages and potential.

However, the existing in-situ adsorbents are not effective for organic pollutants.

(Josefsson et al., 2012)

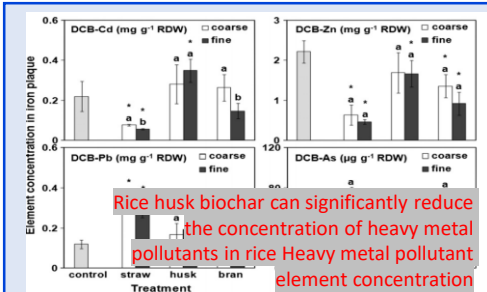
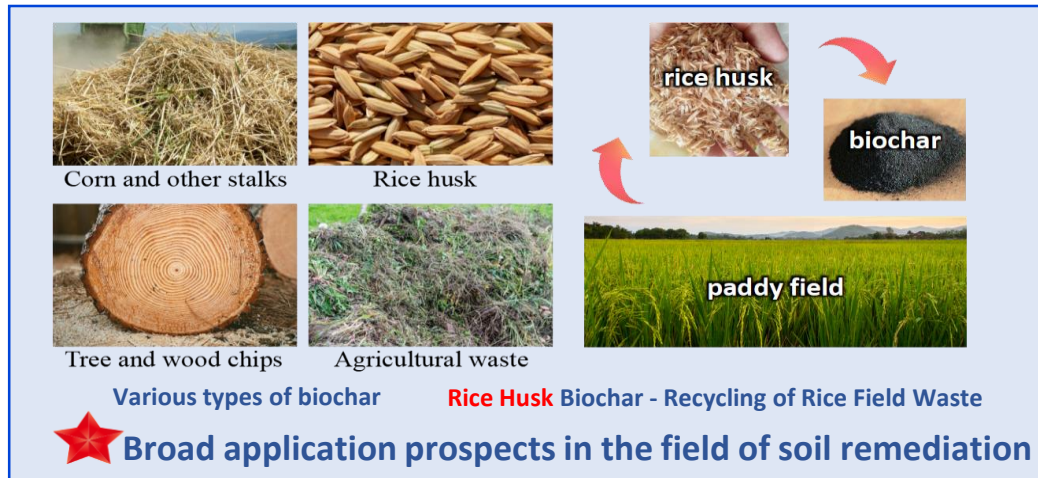
Benthic bioturbation by benthic fauna reduced the remediation efficacy of in situ active mulching techniques.

In conclusion: Finding suitable adsorbents is the key to inhibiting the release of PAHs from paddy field sediments in the cold region.

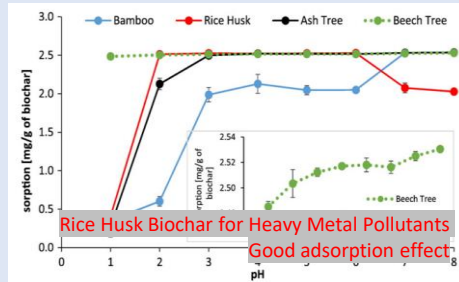
Technical Optimization



- Research on biochar in pollution remediation

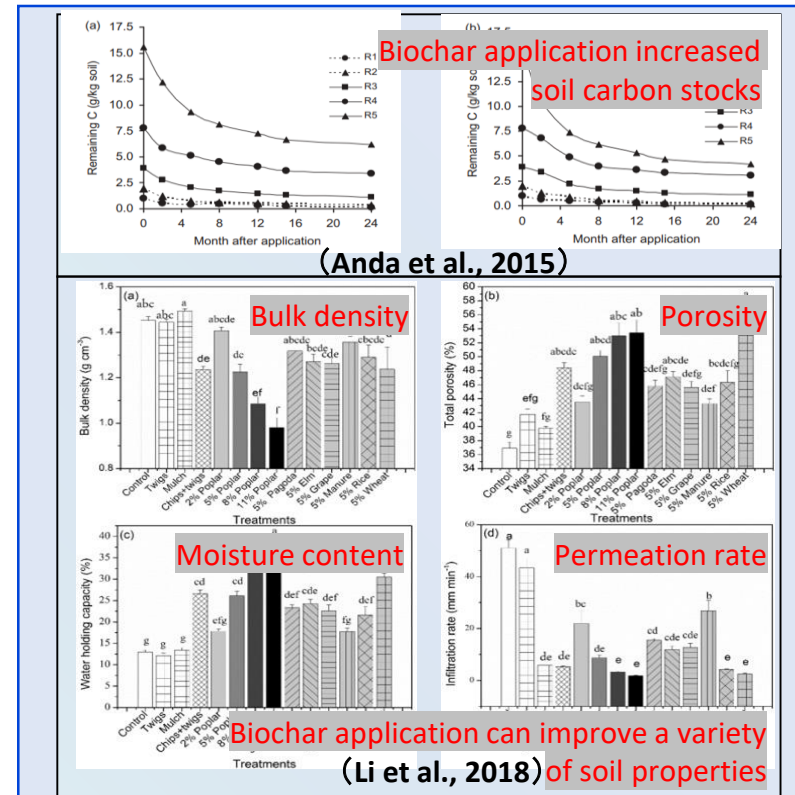


(Zheng et al., 2012)



(Soudek et al., 2017)

Due to its adsorption capacity, **rice husk biochar has better application value in situ remediation of contaminated soil**. Existing studies focused on heavy metals, and fewer studies on organic contaminated soils such as PHE.



Soil application of biochar improves soil properties (pH, moisture, microbial load, and activity), increases soil fertility and crop yields, increases carbon sequestration time, and reduces CO₂ emissions.



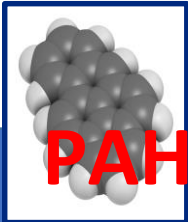
Existing problem

1



It has been shown that benthos disturbance can cause the **structure and properties** changes of the sediments. However, the current research on bioturbation is centered on the **disturbance of benthic organisms** in large ecosystems such as oceans, estuaries, and lakes. **Rice-crab farming has not been adequately studied as a small ecosystem with bioturbation.**

2



PAHs

Currently, most current studies on the remediation of PAHs contamination in the environment by straw biochar have been conducted in water and soil environments. Sediment systems are more complex and variable than individual water and soil environments, **and the remediation of PAHs in the sediments of paddy systems is of substantial significance for food security and human health reasons.**

3




Little research has been reported on the remediation of PAHs contamination in sediments of integrated rice farming systems containing bioturbation conditions, **the effect of biochar addition on the biological effectiveness of PAHs in integrated rice farming systems is unclear. The mechanism of this release is unknown and needs to be explored.**

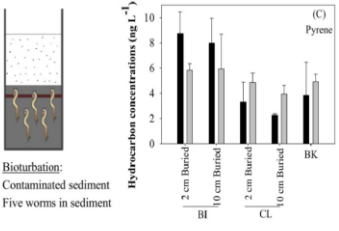
Existing problem




- The mechanism of this release is unknown and needs to be explored.



(Qin et al., 2014)
Various bioturbation effects

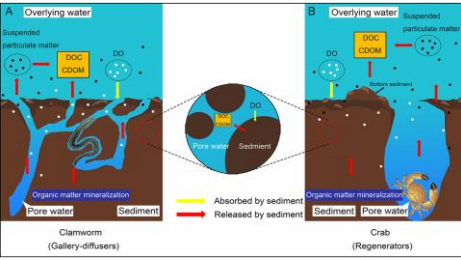


(Tian et al., 2019)
Disturbance of sandworms in double-toothed enclosures (marine)

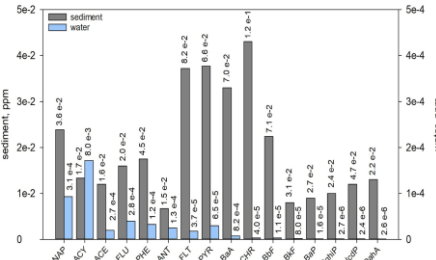


(Mermillod-Blondinet al., 2008)
Perturbation of tubular worms (freshwater)

Significant effects of bioturbation effects on PAHs release



(Nie et al., 2021)
Related studies have shown that bioturbation significantly enhanced DOC concentration in overlying water, which has a strong adsorption capacity for organic matter, thus promoting the release of PAHs from sediments.



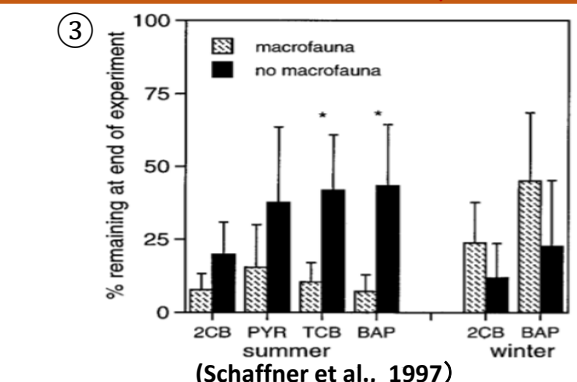
(Jesus et al., 2022)

Facilitating release

③

Release inhibition

?



(Schaffner et al., 1997)
Some studies have shown that bioturbation **inhibits** the release of PAHs from sediments

1. It indicates that the release of PAHs from sediments in freshwater ecosystems is influenced by a variety of environmental factors, such as sediment, benthic fauna.

2. The mechanism of this release is unknown and needs to be explored.

Research Design

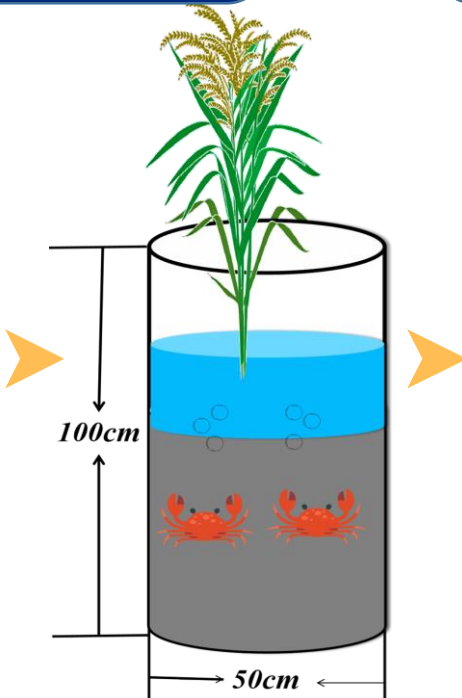


Release of benthic phenanthrene(PHE) by bioturbation



Indoor microcosmic system

Microcosmic experimental design

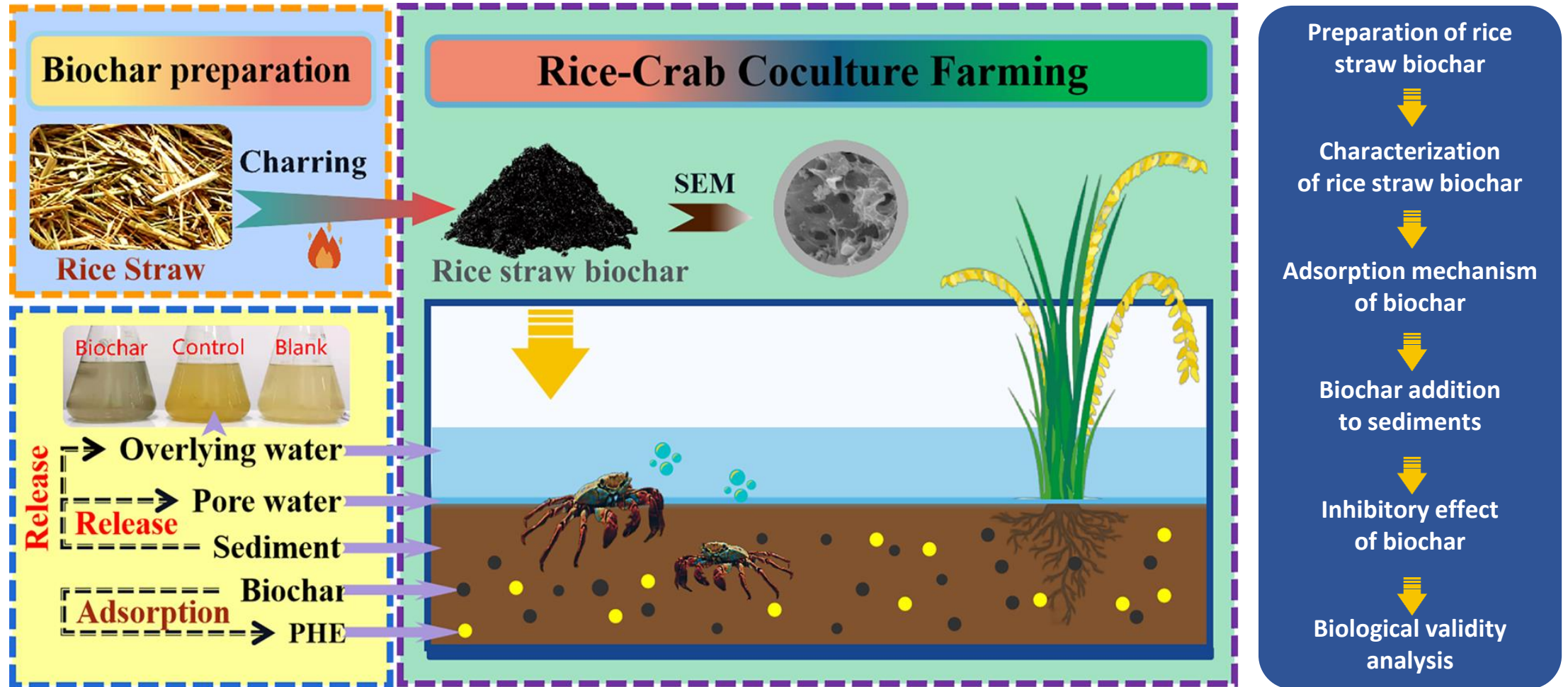


microcosmic device

Two treatment groups, the control group and the crab disturbance group were set up. The experimental period was 30 d. Overlying water samples with a volume of 1 L were collected every 5 d. The physicochemical properties of the overlying water, such as dissolved phenanthrene(PHE), particulate phenanthrene(PHE), total suspended solids (TSS) and dissolved organic carbon (DOC), were measured.

The microcosmic system was a cylindrical device with a diameter of 50 cm and a height of 100 cm. The device contained rice, Chinese mitten crab, overlying water, and sediments.

Research Design



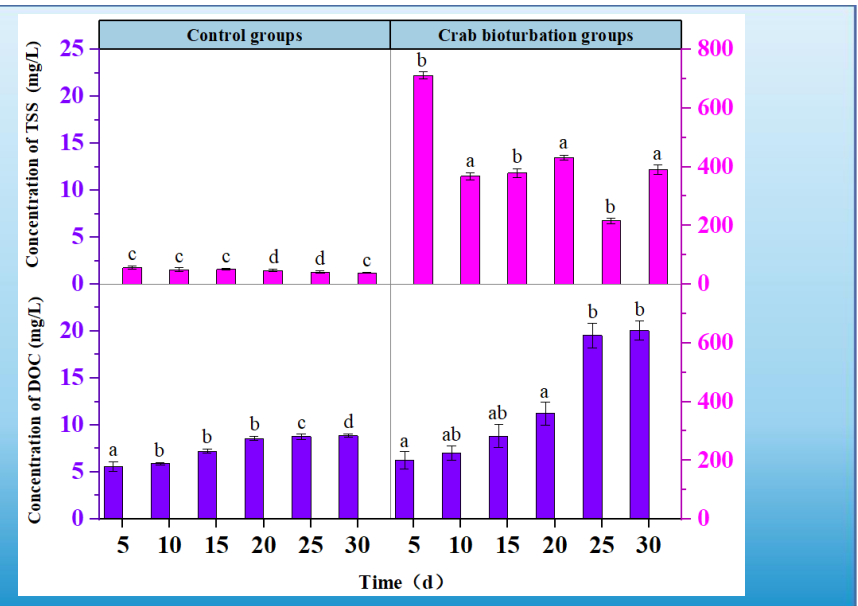
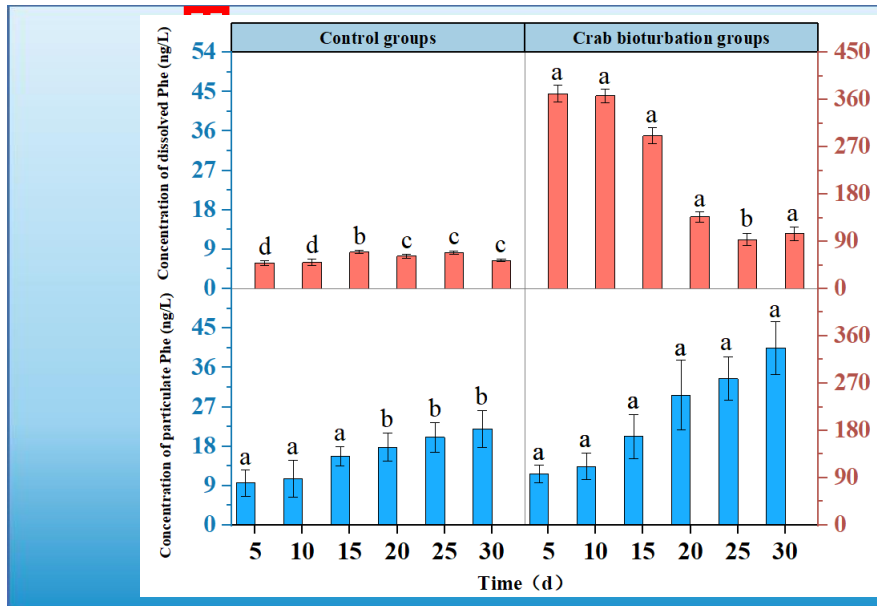
Research result 1: Effect of bioturbation on the release of PHE from sediments



Effects of bioturbation on the release of PHE from the sediments



Changes in physicochemical properties of TSS and DOC in overlying water



Results : The release concentrations of PHE in the particulate and dissolved states of the sediments in the crab bioturbation groups were 5.3 and 1.8 times higher than those in the control group without crab bioturbation, respectively.

Analyze : Bioturbation significantly contributed to the release of PHE from the sediments to the overlying water.

Results : TSS and DOC concentrations in the overlying water of the crab bioturbation group were 18.3 and 3.6 times higher than those of the control group without crab bioturbation, respectively.

Analyze : Bioturbation significantly contributed to TSS and DOC concentrations increase in overlying water.

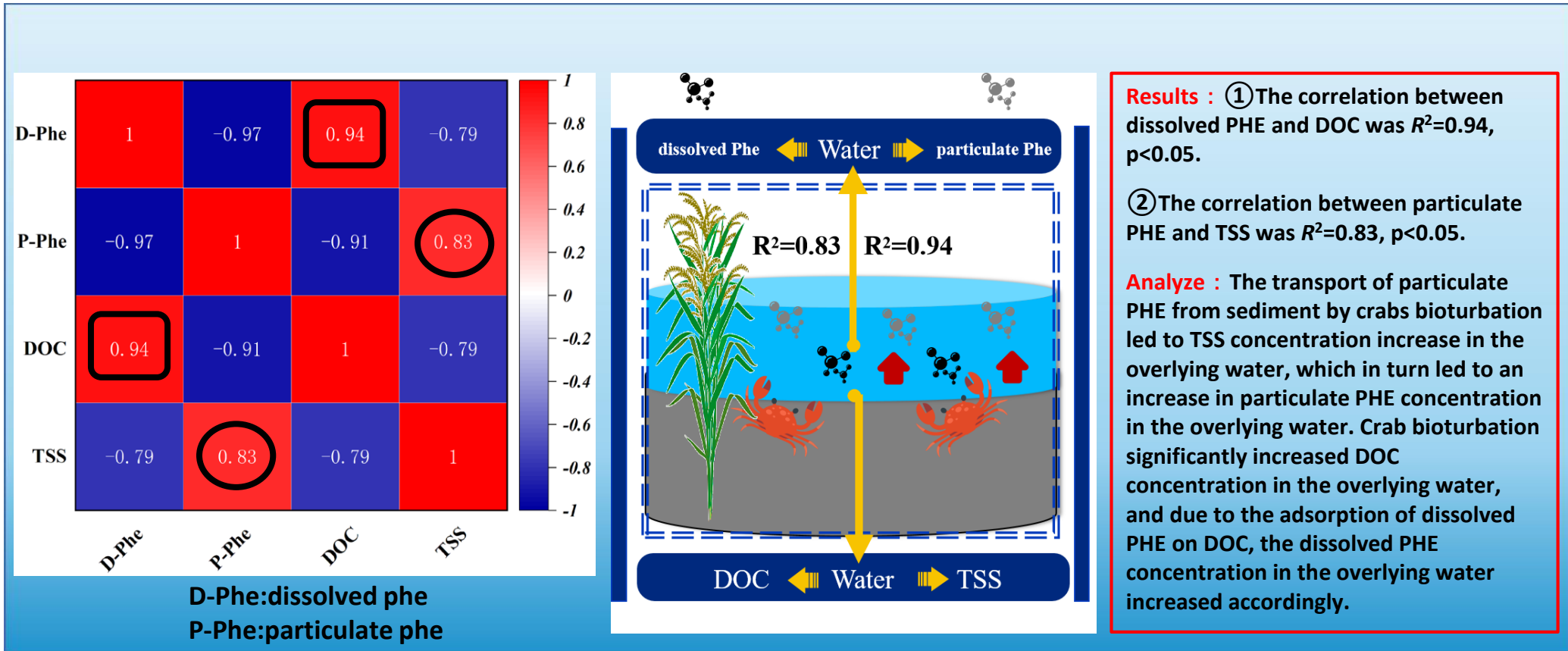
Research result 1: Effect of bioturbation on the release of PHE from sediments



Pearson correlation analysis ($p < 0.05$)

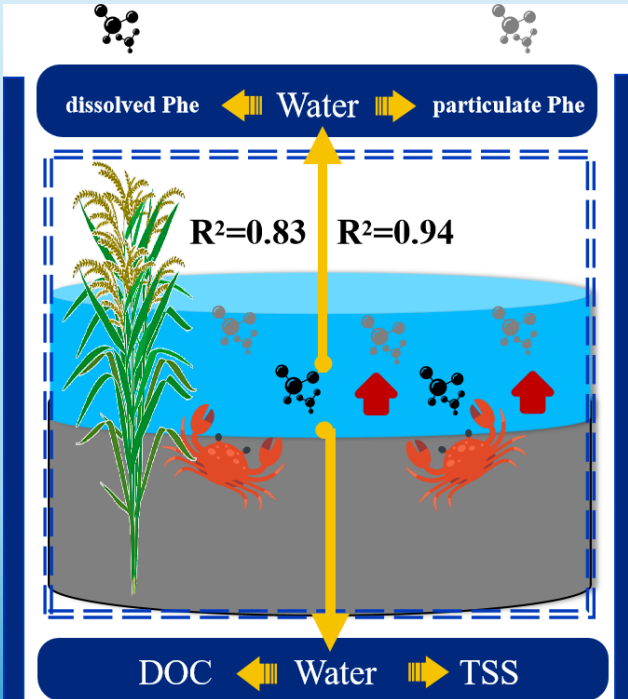


PHE mechanistic analysis of upward water releases



D-Phe	1	-0.97	0.94	-0.79
P-Phe	-0.97	1	-0.91	0.83
DOC	0.94	-0.91	1	-0.79
TSS	-0.79	0.83	-0.79	1
	D-Phe	P-Phe	DOC	TSS

D-Phe:dissolved phe
P-Phe:particulate phe



Results :

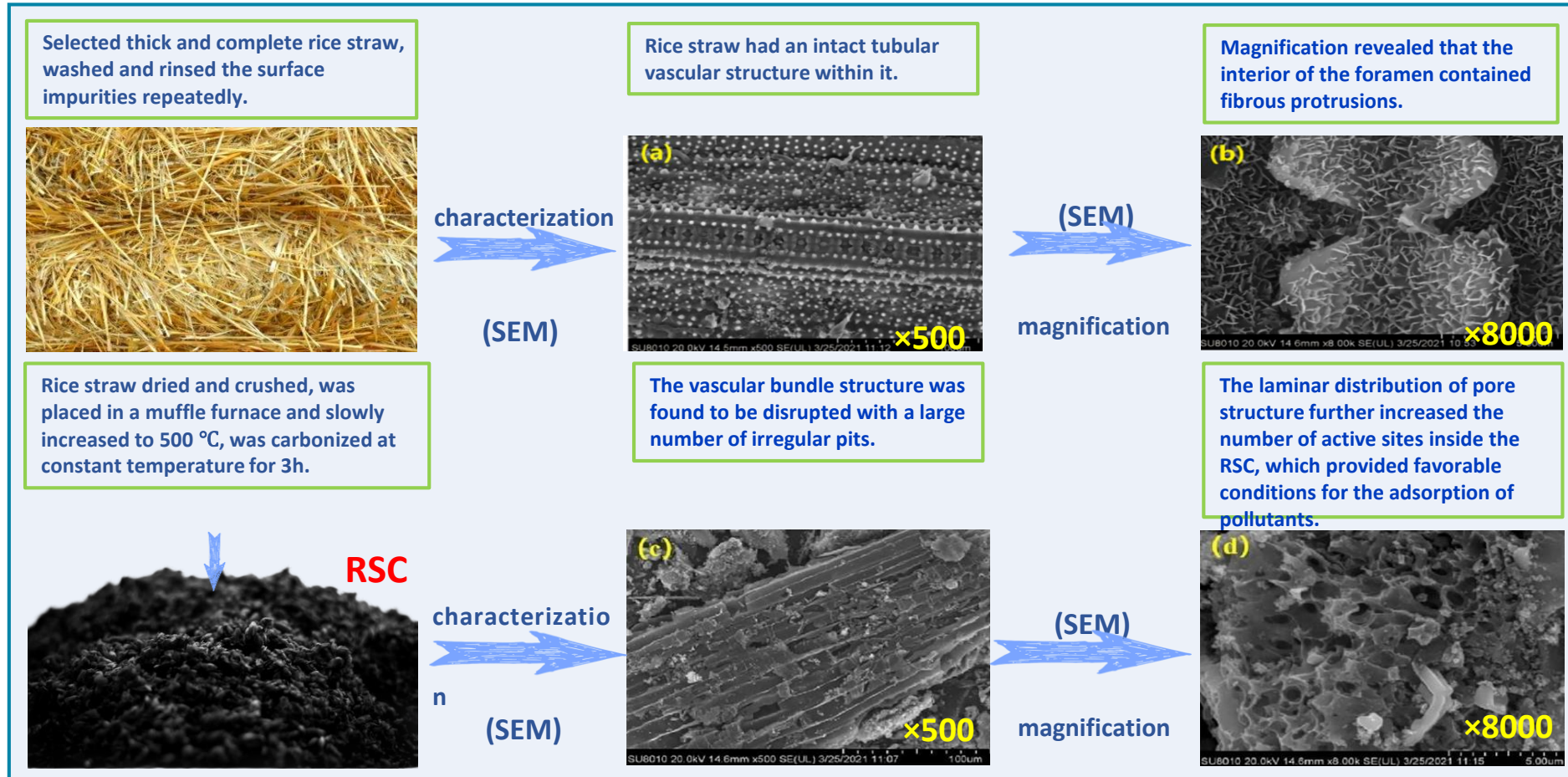
- ① The correlation between dissolved PHE and DOC was $R^2=0.94$, $p < 0.05$.
- ② The correlation between particulate PHE and TSS was $R^2=0.83$, $p < 0.05$.

Analyze : The transport of particulate PHE from sediment by crabs bioturbation led to TSS concentration increase in the overlying water, which in turn led to an increase in particulate PHE concentration in the overlying water. Crab bioturbation significantly increased DOC concentration in the overlying water, and due to the adsorption of dissolved PHE on DOC, the dissolved PHE concentration in the overlying water increased accordingly.

Research result 2: Characterization of PHE adsorption on rice straw biochar



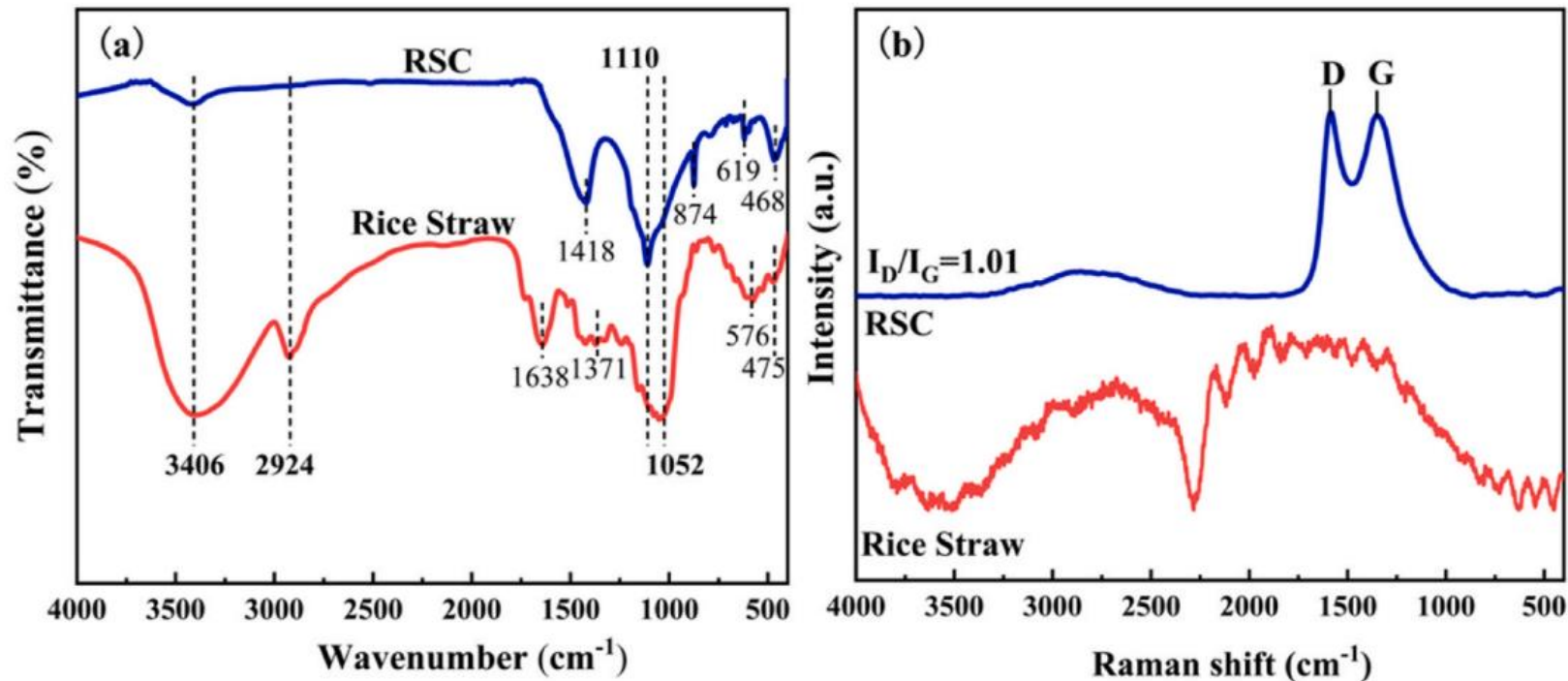
Explored the surface topography of biochar



Research result 2: Characterization of PHE adsorption on rice straw biochar



Explored Fourier transform infrared spectra and Raman spectra



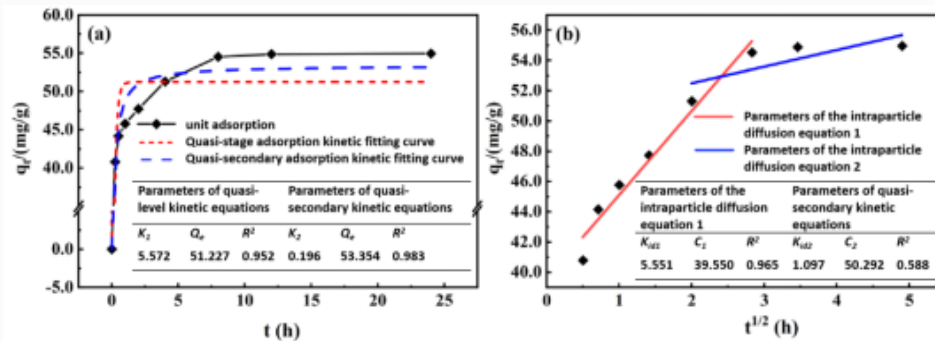
FTIR showed that the high degree of carbonization and high aromaticity of RSC can also improve the carbon content of soils and the stability of biochar in soils, improving soil nutrient conditions and thus promoting crop growth.

Research result 2: Characterization of PHE adsorption on rice straw biochar



Explored Biochar Adsorption Characterization

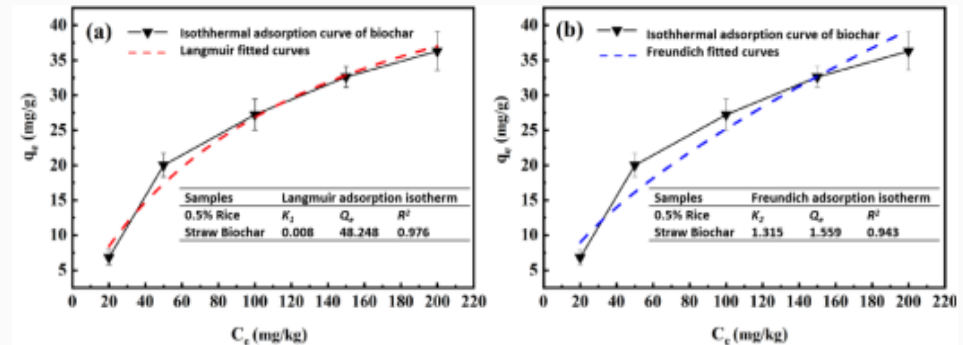
Adsorption kinetic model fitting for PHE adsorption from sediment by rice straw biochar



Results: PHE adsorption by rice straw biochar increased rapidly in the first 3h, then the adsorption rate slowed down after 6h, and basically reached the adsorption equilibrium after 15h. The coefficient of the second-order kinetic model ($R^2=0.983$) was much larger than that of other models.

Analyze: The results showed that the adsorption kinetic process in this experiment conformed to the **quasi-secondary kinetic model**, indicating that the **adsorption** process was mainly dominated by **chemisorption**.

Adsorption isothermal model fitting for PHE adsorption from sediment by rice straw biochar



Results: PHE adsorption by rice straw biochar increased with its initial concentration, and the **Langmuir** isothermal adsorption model ($R^2=0.976$) was better fitted for the PHE adsorption process by rice straw biochar compared to the Freundlich isothermal adsorption model ($R^2=0.943$).

Analyze: It was shown that the adsorption mechanism of PHE by rice straw biochar was complex and dominated by **monomolecular layer adsorption**.



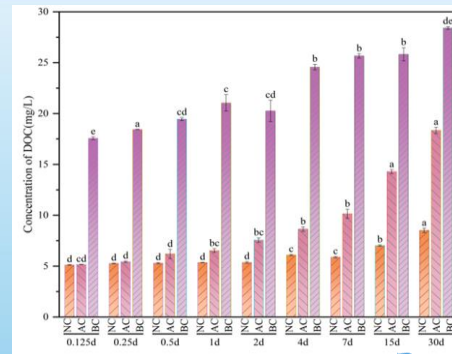
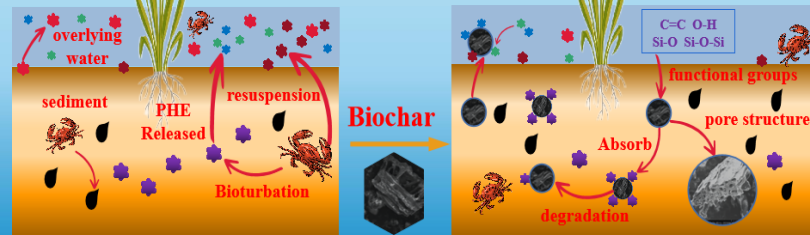
Research result 3: Changes in dissolved PHE content in overlying water

Explored changes in dissolved PHE content in overlying water

🌿 PHE (Sediments state) 🌿 PHE (Particle state)

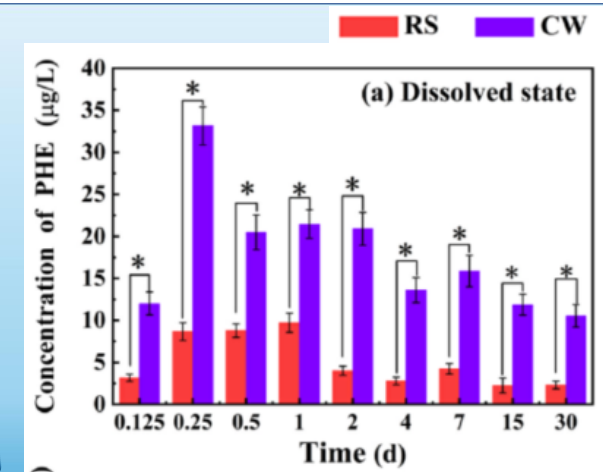
🌿 DOC 🌿 TSS 🌿 TOC 🌿 PHE (Dissolved state)

- Rice straw biochar (RSC) at a concentration of 0.5% (W/W) was added to all sediments and mixed uniformly (called RS)
- sediments were not spiked with any amount of biochar (called CW)



Change in DOC content in overlying water

significant correlation



Changes in dissolved PHE content in overlying water in biochar-added vs. biochar-free systems

★ The concentrations of dissolved PHE in the CW and RS ranged from 10.55 to 33.16 µg/L and 2.25–9.73 µg/L during the experimental period, respectively. **The addition of biochar caused a significant reduction in the dissolved PHE content (P < 0.01) compared with the CW treatment.**

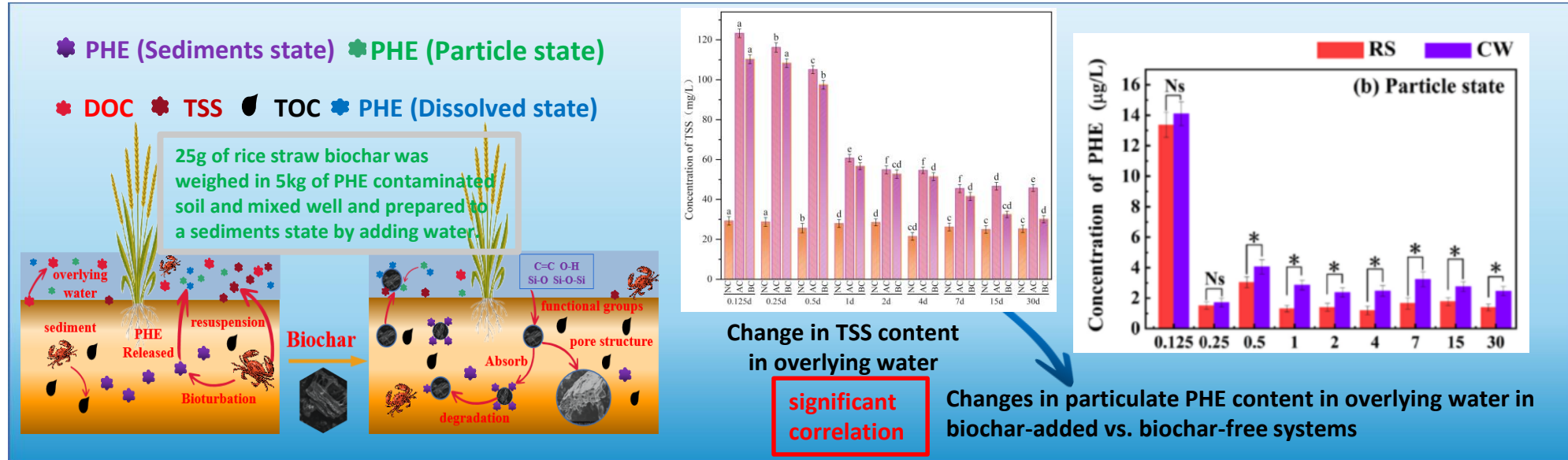
📊 The removal rate of dissolved PHE by biochar exceeded 50% throughout the experimental cycle, and the content of dissolved PHE decreased by 78.03% after 30 d of remediation. **Results**

★ Bioturbation introduced a portion of the biochar particles from the sediments into the water column, and the freely dissolved PHE in the water was absorbed by the reactive sites provided by the large number of pore structures on the surface of the biochar, which reduced the level of contaminants in the overlying water. The activity of crabs can produce a large amount of dissolved organic matter (DOC), and the presence of DOC contributed to a reduction in the PHE content of water. **Analyze**



Research result 4: Changes in particulate PHE content in overlying water

Explored changes in particulate PHE content in overlying water



★ The particulate PHE content in RS was **significantly reduced** compared to CW. ($P < 0.05$) The particulate PHE in the overlying water of RS treatment was reduced by 43.72% compared to CW.

📡 RSC added to sediments can effectively inhibit the migration and release of PHE from the sediment to the overlying water.

Results

★ Crab bioturbation caused an increase in TSS in the overlying water ($P < 0.05$). the suspended particulate matter had a strong adsorbing effect on particulate PHE to **reduce the content of PHE in the overlying water**.

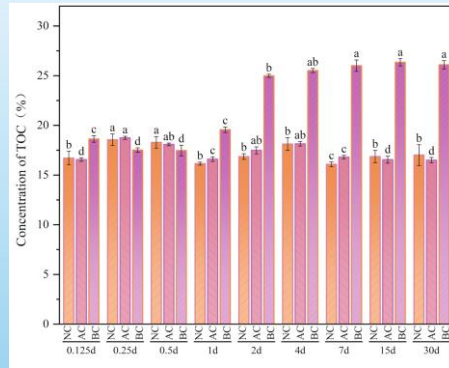
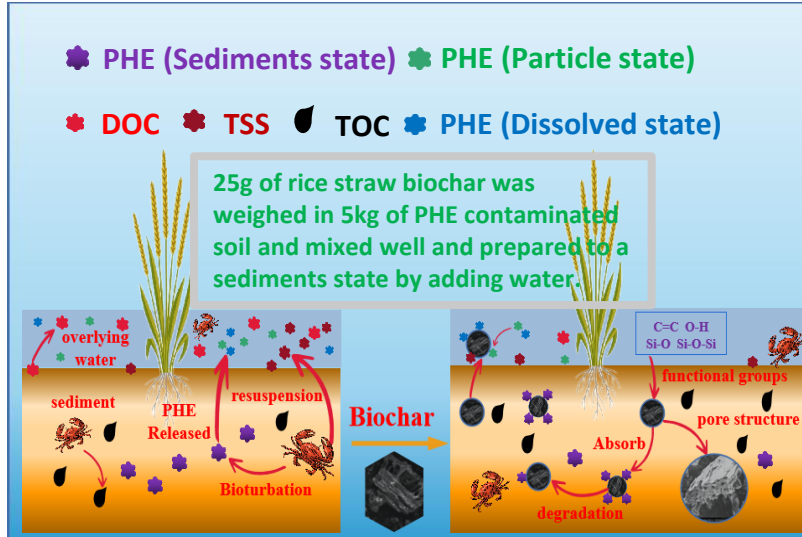
📡 RSC particles adsorbed on the sediment surface are also introduced into the overlying water by resuspension due to bioturbation, and the biochar separated from the sediment surface could adsorb particulate PHE to **reduce the content of PHE in the overlying water**.

Analyze



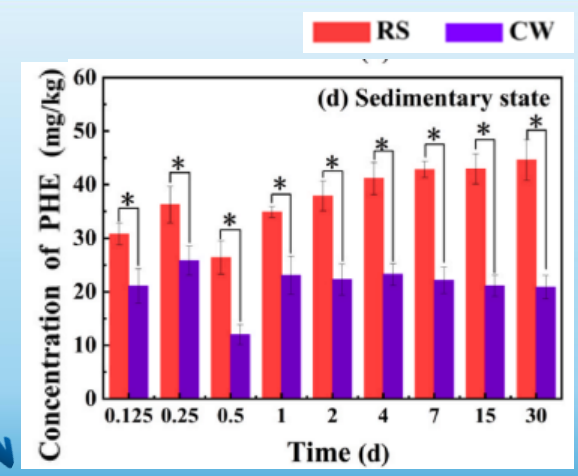
Research result 5: Changes in PHE content in the sediments

Explored changes in PHE content in the sediments



Changes in TOC content in the sediments

Significant negative correlation



Changes in PHE content in the sediments of rice straw biochar-added versus no biochar system

★ No statistically significant difference in PHE content in the sediments of CW was found at the beginning and after 30 d, indicating a relatively slow decay process for PHE in the sediments in the microcosm system.

⬡ The content of PHE in the RS sediments increased by 40.41%–119.62% within 30 d compared with the CW sediments, indicating that the addition of biochar had a significant fixation effect on the PHE in the sediments.

Results

★ RSC in the sediment adsorbed the PHE in the sediment through its complex pore structure and developed specific surface area.

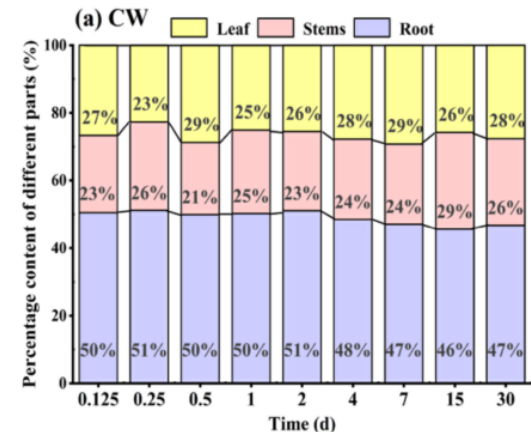
The PHE content in the sediment was significantly negatively correlated with the TOC content in the sediment, showing that rice biochar can be used as a metabolic carbon source for microorganisms, promote the growth of PAH-degrading strains, and accelerate PHE degradation in sediment.

Analyze

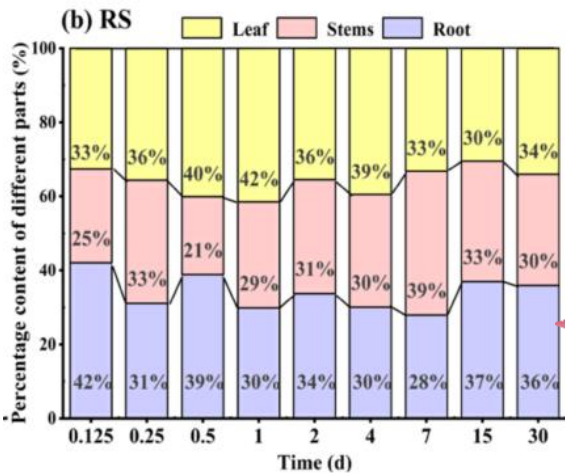


Research result 6: changes in PHE content in plants

Explored changes in PHE content in plants

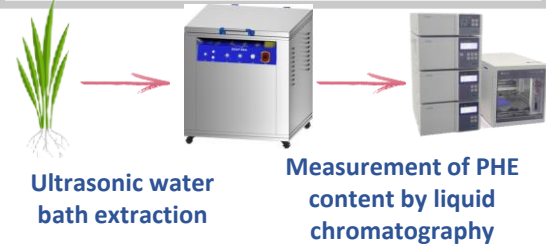


Percentage of PHE in each part of rice in CW treatments



Percentage of PHE in various parts of rice in RS treatments

- 1. After rice growth entered the **tillering stage**, rice plants with straight stalks and rapid growth were selected as experimental rice.
- 2. Sampling times were **3h, 6h, 12h, 24h, 2d, 4d, 7d, 15d and 30d** after the start of the experiment, and **three** sets of parallel samples were set for each treatment at each sampling point.



1. However, there was a **large difference** between the contents of PHE in rice from the two treatment groups. The proportion of PHE in the stems and leaves of rice in RS increased by **2%–15%** and **4%–17%**, respectively, while the proportion in the roots showed a significant decrease ($P < 0.01$).

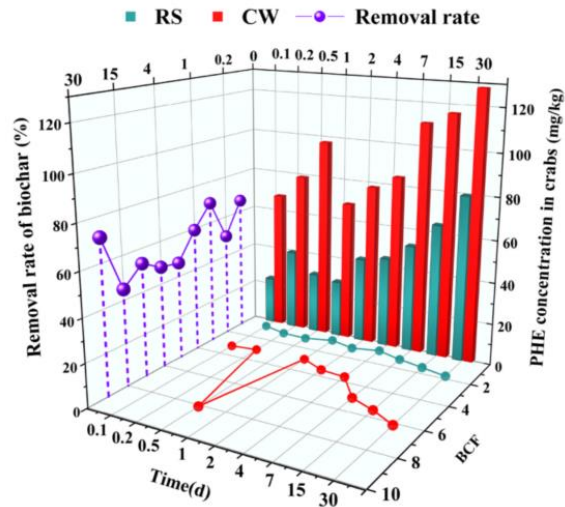
2. The inhibitory effect of biochar on plant uptake of organic pollutants may be due, on the one hand, to the fact that the biochar reduced the concentration of **bioavailable PHE** in the soil, and on the other hand, the loose and porous structure of biochar provided a suitable living environment and shelter for a variety of **PAHs-degrading bacteria** in the soil.

3. The result confirmed that the transfer of PHE from sediments to plant roots was **greatly limited** due to the addition of **biochar**. It was still unknown how biochar interfered with and inhibited the uptake and **translocation of PAHs** in different parts of the plant, but the effect of biochar addition on the alteration of soil properties and the signaling between interroot microorganisms cannot be excluded.



Research result 7: Changes in PHE levels in crabs

Explored changes in PHE levels in crabs

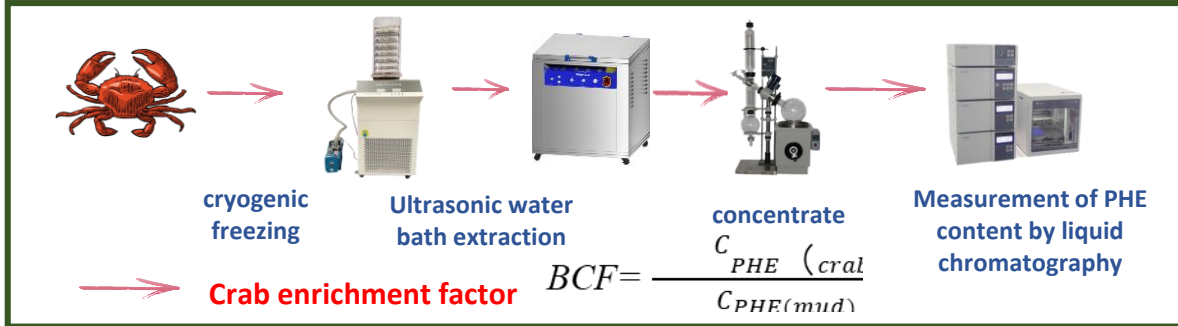


The crabs were destructively sampled at 3 h, 6 h, 12 h, 24 h, 2 d, 4 d, 7 d, 15 d and 30 d in the system.

1. Although the PHE concentration in the artificially polluted sediments in this study was high, the crabs exposed to the sediments showed a high survival rate (>86%), which may be because crabs in the larval stage were more tolerant to PAHs. However, a significant decrease was observed in the biological activity of the crabs in both CW and RS after 30 d of being exposed to PHE in the sediments.

2. The total uptake of PHE and the BCF in crabs in RS was significantly lower ($P < 0.01$). After 30 d of biochar amendment, the enrichment of PHE by crabs in RS was **38.17%** lower compared with CW, indicating that the addition of straw biochar effectively inhibited the uptake of PHE from the sediment by crabs.

3. The process of PHE immobilization by biochar in the sediments also reduced the ability of PHE to be released into the water in a **bioavailable state**, which blocked the uptake of PHE from the water and sediments by crabs during **respiration and feeding activities**.





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Inhibition of polycyclic aromatic hydrocarbon (PAHs) release from sediments in an integrated rice and crab coculture system by rice straw biochar

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ABSTRACT

An persistent organic pollutants that are widely present in the environment and that pose substantial threats to human health, polycyclic aromatic hydrocarbon (PAHs) have received much attention from research around the world, but few studies have investigated the PAHs contamination of sediments in rice and crab coculture farming systems. In this study, rice straw biochar was prepared from rice straw, an abundant agricultural waste product, under nitrogen-limited conditions at 300 °C. The rice straw biochar was characterized by scanning electron microscopy (SEM), specific surface area analysis (SSA), Fourier transform infrared spectroscopy (FTIR), and Raman Spectroscopy. The effect of using the straw biochar as a substrate on the inhibition of the release and migration of PAHs from sediments in a rice and crab coculture system and the biological effects of PAHs within the system were studied for the first time. The results showed that rice straw biochar has a rich pore structure and high specific surface area, and the high-temperature charring method utilized in this study improved the charring and aromaticity of rice straw. The addition of rice straw biochar increased the immobilization of PAHs in sediments by 40–67%, 11–122% and reduced the migration of PAHs from the sediments to the overlying water, which reduced the PAHs content of the pore water at the sediment-water interface by 11.47%, 56.75% and reduced the PAHs content of the overlying water in the dissolved and particulate forms by 54.07%, 61.20%, and 1.09%, 52.26%, respectively. The addition of rice straw biochar reduced the biological effectiveness of PAHs in the system, and the content of PAHs in crabs and rice decreased by 28.37–69.55% and 52.22%–97.04%, respectively. In addition, rice straw biochar changed the distribution of PAHs in rice, increasing the PAHs content of the stalks and leaves of rice by 2%, 15% and 4%, 17%, respectively, while the PAHs content of the rice was reduced by 9%–20%. These results confirm that rice straw biochar can effectively inhibit the migration and release of PAHs from sediments and reduce the bioavailability of polycyclic aromatic hydrocarbon (PAH) in integrated farming systems. The results provide theoretical and technical support for the study of waste water utilization and remediation of PAHs pollution.

1. Introduction

An integrated rice farming model is an efficient integrated farming model that can effectively promote sustainable agricultural development, and it is designed based on the ecological characteristics of rice combined with the biological characteristics of fish, crabs, and other aquatic species (Song et al., 2021). This farming model can take advantage of the synergistic effects of mutually beneficial symbiotic material cycles between species within a given spatial area, and with the aim of enabling effective rice production and aquaculture. It can also reduce the use of chemical fertilizers and pesticides and make more rational use of natural resources (Fu et al., 2019; Zhenxing et al., 2021). Chinese mitten crabs (*Decapoda reticulata*) is a crab in an important aquatic animal in China. After the initial success of the rice fish

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Remobilization and bioavailability of polycyclic aromatic hydrocarbons from estuarine sediments under the effects of *Nereis diversicolor* bioturbation^a

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ABSTRACT

The effects of *Nereis diversicolor* bioturbation on the remobilization and bioavailability of polycyclic aromatic hydrocarbons from estuarine sediment were determined after 60 d in a laboratory experiment. The release fluxes and mass transfer coefficients showed that bioturbation by *N. diversicolor* can lead to a significant remobilization of polycyclic aromatic hydrocarbons (PAHs) from estuarine sediments. Bioturbation enhanced the release of PAHs from sediment to water by accelerating the transport of sediment particles to the sediment-water interface followed by PAHs desorption to the water. The bioavailability of PAHs was described by SMAD-sediment accumulation factors (SAAF). The SAAF of low molecular weight PAHs with bioturbation was significantly higher than that of PAHs without bioturbation, and there were no significant variations in high-molecular-weight PAHs. Our results revealed that *N. diversicolor* bioturbation significantly increased PAHs release from sediment to water but only increased the bioavailability of low-molecular-weight PAHs.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous contaminants in the environment, especially in the sediments of estuaries throughout the world, and they are of great interest due to their toxic effects on the environment (Iqbal and Ullrich, 2018). Once PAHs enter a water body, they are preferentially associated with suspended particulate matter and then precipitate to the bottom sediment (Sun et al., 2017). Contaminated sediment can release these substances and thereby act as a secondary source of PAHs to the water column (Ding et al., 2016; Mustajärvi et al., 2017). Several processes, such as mechanical water mixing, wave action, tides, and wind, have been demonstrated to significantly affect on the release of PAHs from sediment to water in the field. Bioturbation is also one of the processes that can lead to direct or indirect PAHs transfer from sediments to water. Bioturbation is a

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Effect of humic acid-modified attapulgite on polycyclic aromatic hydrocarbon adsorption and release from paddy soil into the overlying water in a rice-crab coculture paddy ecosystem and the underlying process

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HIGHLIGHTS

- Modified attapulgite was prepared by hot acid modification with humic acid.
- Crab bioturbation disrupted the Phe distribution balance between paddy soil and overlying water.
- The adsorption efficiency of released dissolved and particulate Phe reached 91.97% and 93.26%.
- HA-ATP and DOC competitively adsorbed dissolved Phe in overlying water.
- HA-ATP immobilized particulate Phe in overlying water during ecosystem.

GRAPHICAL ABSTRACT

ABSTRACT

Humectone (Phe), a typical polycyclic aromatic hydrocarbon (PAH) pollutant, poses an enormous safety risk to rice-crab coculture (RC) paddy ecosystem. In this study, humic acid-modified purified attapulgite (HA-ATP) with a complex structure was successfully fabricated to absorb Phe released from paddy soil to overlying water in RC paddy ecosystem in Northeast China. The maximum crab bioturbation intensifies the dissolved Phe and particulate Phe were 44.83mg/L, 1.00[±]0.40 and 24.23mg/L, 1.00[±]0.24, respectively. The higher concentration of dissolved Phe released from paddy soil to overlying water due to crab bioturbation reached 80.83mg/L, while the corresponding concentration of particulate Phe reached 207.36mg/L. The dissolved organic carbon (DOC) and total suspended solid (TSS) concentrations in overlying water increased correspondingly and were strongly correlated with dissolved Phe and particulate Phe concentrations, respectively (P² = 0.80). When 0% HA-ATP was added to the surface layer of paddy soil, the efficiency of the adsorption of Phe release was 24.00%–26.38% for particulate Phe and 89.99%–91.97% for dissolved Phe. Because HA-ATP has a large adsorption pore size (11.33 nm) and surface area (824.43m²/g) as well as many HA functional groups, it provided multiple hydrophobic adsorption sites for dissolved Phe, which was conducive to competitive

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ABSTRACT

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