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RESEARCH

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# The adsorption effect of aquaculture residual feed on heavy metals and its impact on microbial metabolism of feed

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**Abstract**

**Background** Intensive and large-scale aquaculture has a high biological density, and a large amount of artificial feed must be provided. As a result, residual feed would be discharged into natural water bodies with water exchange. Due to heavy metal pollution in surface water, residual feed may adsorb heavy metals in the water, affecting the subsequent microbial degradation of feed and the restoration of the water environment. The combined ecological impacts of these two types of pollution urgently need to be explored.

**Results** This study explored the adsorption effect of residual feed on heavy metals and its impact on microbial degradation of residual feed. The results showed that both the insoluble component (mainly fiber, ash, and ether extract, abbreviated as FAE) and soluble component (soluble protein and starch) of the feed had adsorption effects on heavy metals. As the salinity increased, the adsorption capacity of $Cu^{2+}$ and $Cd^{2+}$ decreased, while the adsorption effect on $Pb^{2+}$ was not changed significantly. The adsorption of heavy metals by residual feed could affect the biological degradation of residual feed. The residual feed could adsorb heavy metals and bacterial cells simultaneously, forming FAE-B(bacteria)-S(starch)-Pr(protein)-metal adsorption complex. This adsorption effect could reduce the negative impact of heavy metals on microbial degradation of residual feed. The simulated degradation experiments in actual water bodies also confirmed this. However, the accumulation of heavy metals in residual feed may bring negative effects, such as being consumed by aquatic organisms and entering the food chain.

**Conclusions** The results of this study provide a basis for the collection and centralized treatment of residual feed in aquaculture, as well as the control and remediation of residual feed pollution in natural water bodies.

**Keywords** Residual feed, Adsorption complex, Feed pollution, Heavy metal

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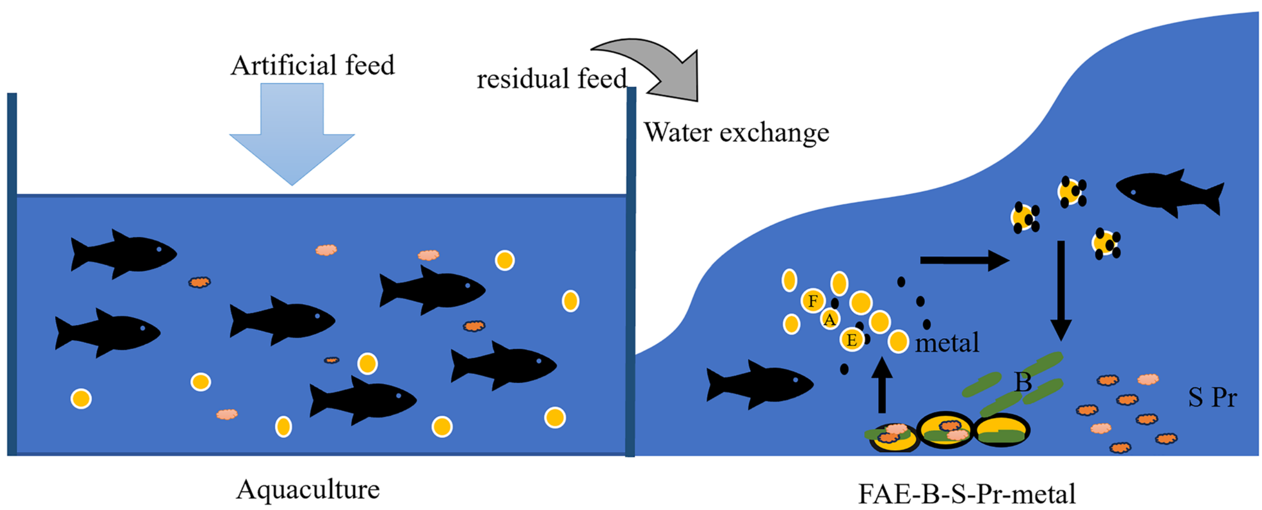
**Graphical Abstract**




## Introduction

In recent years, the aquaculture industry has witnessed rapid development. The industry is formulating a high-density, high-yield intensive farming model (Chang et al. 2020). High-density farming enables cultivating more organisms within limited water area, which significantly increases the output to meet the growing demand. Furthermore, the high-density intensive farming model can reduce the land required for aquaculture, save labor, reduce production costs, and improve economic efficiency (Chen and Gao 2023; Wang et al. 2023).

However, to achieve high-density intensive farming, natural feed in waters is insufficient, and artificial feed must be provided. According to reports, about 5–30% of the existing feed cannot be consumed by animals and becomes residual feed (Fu 2019; Opstvedt et al. 2003). According to the field investigation of marine aquaculture farms in Qinhuangdao City of China, the unused feed accounts for approximately 20–30% of the total feed. Residual feed contains high levels of nutrients such as protein and starch. Excessive amount of these nutrients in localized waters can lead to eutrophication (Gichana et al. 2018). There is a significant correlation between fishing production and the emission of pollutants (Braña et al. 2021). One of the main causes of the deterioration of the aquatic environment in aquaculture waters is residual feed (Kong et al. 2023).

The residual feed is often directly discharged into natural waters with water exchange, which will be purified and degraded by natural water microorganisms (Zheng et al. 2022). According to Li et al. (2017), during the process of seawater aquaculture, the particulate matter on the wall and bottom of ponds would be directly discharged into the sea when manually changing water and cleaning the fish pond. These particulate matters mainly include residual feed and excrement, which can be decomposed by microorganisms in the seawater. Specialized microbial preparations have also been widely accepted to treat the contamination of aquaculture effluents (Ahmed and Turchini 2021). Microorganisms in natural waters and specialized microbial preparations can utilize organic matter in residual feed as a nutrient source and decompose it, but the effectiveness of such methods depends on the activity of microorganisms.

Proteins, carbohydrates, and other substances rich in artificially-prepared feed are heavy metal affinity substances (Hu et al. 2010). With the intensification of industrial pollution and the increase of agricultural activities, surface water often suffers from varying degrees of heavy metal pollution (Yin et al. 2015; Kumar et al. 2019). Even if the heavy metal concentration is low, it may cause negative effects on microorganisms due to the adsorption and enrichment of heavy metals by residual feed. As is well known, heavy metals have a certain toxic effect on microorganisms. Jaafar (2020) reported that $Pb^{2+}$ and $Cd^{2+}$ could inhibit microbial growth. Foulkes (2000) pointed out that heavy metals might cause structural changes of microorganisms. Narwal et al. (2023) showed that heavy metals could bind to enzymes in microorganisms, interfering with their catalytic activity. Morales et al. (2016) proposed that $Cd^{2+}$ and $Ni^{2+}$ could lead to the breakage, damage, and denaturation of DNA and RNA strands. Although microorganisms are partially tolerant to heavy metals, when the concentration of heavy metals exceeds a certain level, they will inhibit normal metabolic activity of microorganisms (Chaturvedi et al. 2015). Akpor et al. (2014) explored the impact of heavy metals on the wastewater treatment systems and confirmed the adverse effects of heavy metals on microbial degradation of organic matter. In summary, there is an urgent need to study whether the adsorption of heavy metals by feed will affect the degradation process of residual feed by microorganisms.

Therefore, this research systematically explored the adsorption effect of residual feed on heavy metals and its impact on the microbial degradation of residual feed. Firstly, the heavy metal adsorption capacity of residual feed was analyzed, and efforts were made to investigate the effects of pH, salinity, and temperature on the adsorption of typical heavy metals by residual feed. Then, the impact of heavy metals adsorption on the microbial degradation of residual feed was analyzed combining the results of the adsorption experiment. Finally, discussion was conducted about the effects of heavy metal adsorption on the microbial degradation of residual feed in natural water bodies. The results of this study can provide a basis for the collection and centralized treatment of residual feed in aquaculture, as well as the control and remediation of residual feed pollution in natural water bodies.

## Materials and methods

### Materials

The chemical agents (produced by Tianjin Chemical Reagent Factory) used in this experiment were all analytical pure. The feed was provided by Hebei Academy of Marine and Aquatic Sciences and was a kind of compound feed for adult sturgeon fish. The feed used in this study was sold feed that met national standards, and it did not contain toxic heavy metals, including lead, mercury, cadmium, etc. The feed was screened with a 40–80 mesh sieve after crushing, and dried at 60 °C in an oven for 24 h to prepare for experiments. The composition of feed is shown in Table S1. The main components included protein, fiber, ash, ether extract and so on. The specific surface area of feed was $1.23 \pm 0.18$ $m^2/g$. The biological reagent used in the experiment was purchased from Sigma Company. *Bacillus licheniformis* (ATCC 11946) was stored in the laboratory.

### Culture medium

The activation medium was composed of 5.0 g/L yeast extract, 10.0 g/L tryptone, 0.1 g/L potassium nitrate. 5 g/L of agar was added to obtain a solid culture medium. The pH of the medium was adjusted to 7.2–7.4. The sterilization temperature of the culture medium was 121 °C, and the sterilization time was 30 min.

### Activation and amplification of the strain

The preserved strain was inoculated into liquid activation medium, and then was cultivated at 37 °C and 180 r/min for 24 h. The bacterial suspension was transferred into a 15 mL centrifuge tube, and centrifuged at 4000 r/min for 10 min. Then the supernatant was discarded and 10 mL of sterile physiological saline was added into the tube. The mixed solution was shaken well to obtain the bacterial suspension. 1 mL of bacterial suspension was transferred into a centrifuge tube and 9 mL of sterile physiological saline was added into the tube. The mixed solution was shaken again. 100 μL bacterial solution was then coated on the solid culture medium and incubated in an incubator at 37 °C for 24 h. After picking the single colony, three streak purification was performed on solid culture medium. The purified strain was preserved at − 20 °C. At the same time, the purified strain was inoculated into liquid culture medium and cultivated for 24 h at 37 °C and 180 r/min. The bacterial solution was then poured into a centrifuge tube and centrifuged at 4000 r/min for 10 min. After rinsing the bacterial precipitation with sterile physiological saline for three times, the bacterial cells were transferred into the sterile physiological saline solution to prepare the bacterial suspension with OD600 of 1.6 for subsequent use.

### The adsorption experiment of heavy metals

To distinguish the adsorption differences of soluble and insoluble components of residual feed on heavy metals, the adsorption effects of the two on heavy metals were tested separately. For each adsorption experiment, two experimental groups were set up. The feed was placed in a conical flask, and then deionized water was added. The feed concentration was 2.5 g/L. The mixture was thoroughly stirred to dissolve. After centrifuging the mixture, the insoluble matters (mainly fiber, ash, and ether extract, abbreviated as FAE) was removed from one of the groups, which was referred to as the group without insoluble components (NO-FAE group, abbreviated as NFAE group). The other group is the group retaining the insoluble components (FAE group). The proportion of FAE in feed (dry weight) was $59.46 \pm 4.90\%$.

Then, 100 mg/L of $CuSO_4$, Cd $(NO_3)_2$, and Pb $(NO_3)_2$ solution was added to the conical flask separately. The conical flasks were placed in a constant-temperature oscillator and shaken at 180 r/min under room temperature and neutral conditions to achieve adsorption equilibrium (about 48 h). Then the concentration of heavy metals in the water sample was measured.

In addition, the effects of pH (4, 5, 6, 7, 8), temperature (25, 30, 35, and 40 °C) and salinity (0.0, 5.0, 10.0, 15.0, 20.0 g/L) on the heavy metal adsorption capacity of feed were investigated. The pH value of the solution was adjusted by 0.1 mol/L NaOH solution and 0.1 mol/L HCl solution. Different masses of NaCl were weighed and heavy metal solutions with different salinity were prepared. The concentration of heavy metal ions was the same as in the previous experiment. The experiments on the adsorption of heavy metals by residual feed were conducted by the same method, and the mass of heavy metals adsorbed by residual feed was calculated. Then, the feed adsorption experiments were conducted under the conditions of 25 °C and 180 r/min, and the adsorption kinetics model fitting was performed. This research used quasi-first-order kinetics, quasi-second-order kinetics, and Elovich kinetics to fit the adsorption process. In addition, the blank experiment was conducted without the addition of feed, and the adsorption experiment was conducted under the same conditions. The concentration of the initial heavy metal ions was the same as in the previous experiment.

#### The microbial degradation of residual feed

Two different types of residual feed wastewater (FAE, NFAE) were prepared using the same method as in Sect. "The adsorption experiment of heavy metals". In practical situations, the adsorption of heavy metals by residual feed and the degradation of residual feed by microorganisms usually occur simultaneously. Therefore, 100 mL residual feed wastewater with $Cu^{2+}$ concentrations of 0.0, 2.0, 5.0, 10.0, 30.0, and 50.0 mg/L was prepared respectively in the conical flasks and sterilized. Then the bacterial suspension was inoculated with the inoculation amount of 3% (v/v). The conical flasks were placed in a constant-temperature oscillator and shaken at 180 r/min, 30 °C. The water solution was sampled every 24 h and the concentration of soluble starch, soluble protein, and bacterial biomass were measured. When determining soluble starch and soluble protein, the solution was centrifuged and the supernatant was detected. The experimental steps for investigating the effects of $Cd^{2+}$ and $Pb^{2+}$on the bacterial degradation of residual feed are the same as those for the $Cu^{2+}$ effect experiment mentioned above. This experimental method was adopted to better simulate the actual situation.

#### Microbial degradation of feed in natural waters

The experimental water was taken from the Yanfei Lake of Yanshan University and the costal seawater of Qinhuangdao City in Hebei Province, China. The lake water and seawater were filtered and stored for use. Four groups of experiments were set up. No heavy metal ions were added to the blank experimental group. The concentrations of heavy metals in the low concentration experimental group were $Cu^{2+}$ (1 mg/L), $Cd^{2+}$ (0.1 mg/L), and $Pb^{2+}$ (0.1 mg/L), respectively. The concentrations of heavy metals in the medium concentration experimental group were $Cu^{2+}$ (5 mg/L), $Cd^{2+}$ (1 mg/L), and $Pb^{2+}$ (1 mg/L), respectively. The concentrations of heavy metals in the high concentration experimental group were $Cu^{2+}$ (10 mg/L), $Cd^{2+}$ (5 mg/L), and $Pb^{2+}$ (5 mg/L), respectively. The mixed solutions containing $Cu^{2+}$, $Cd^{2+}$, and $Pb^{2+}$ and filtered water were prepared. Then, the residual feed was added to the heavy metal mixed solution. The purpose of the experiments was to evaluate the impact of varying levels of heavy metal ion concentrations on degradation. The degradation effects of microorganisms in the lake water and seawater on residual feed were tested separately. The content of soluble starch, soluble protein, and ammonia nitrogen in the water sample was measured every 24 h.

#### Analytical methods

The concentration of heavy metals was measured by an atomic absorption spectrophotometer (Shimazu, AAS06800). The amounts of heavy metals adsorbed by residual feed (adsorption capacity) can be calculated by

$$q = \frac{(C_0 - C_e)V}{M} \tag{1}$$

where $q$ is the amounts of heavy metals adsorbed by residual feed (mg/g); $C_0$ is the concentration of $Cu^{2+}$, $Cd^{2+}$ and $Pb^{2+}$ in the solution before adsorption (mg/L); $C_e$ is the concentration of $Cu^{2+}$, $Cd^{2+}$ and $Pb^{2+}$ in the solution after adsorption (mg/L); $V$ is the solution volume (L); $M$ is the quality of the feed (g).

The degradation efficiency or adsorption efficiency was calculated as follows:

Degradation or adsorption efficiency = (concentration before degradation or adsorption − concentration after degradation or adsorption)/concentration before degradation or adsorption × 100%.

The soluble starch content was measured using iodine colorimetric method (French et al. 1973). The soluble protein content was determined by the Coomassie brilliant blue method (Li et al. 2000). The microorganism concentration OD600 was measured using a spectrophotometer. The Nessler's reagent spectrophotometry was used to determine the concentration of ammonia nitrogen in residual feed wastewater (Lin et al. 2014). In order to analyze the adsorption of heavy metals by residual feed on bacterial cells during the degradation of residual feed by *Bacillus*, a scanning electron microscope (COXEM, EM-30AXN) equipped with EDS spectrum was used to observe the morphology of residual feed in wastewater and determine the adsorption status of bacteria and the heavy metal. Feed was also observed as the blank control group.

### Statistical analysis

All the mentioned experiments were repeated three times, and the average data were obtained. One-way ANOVA was used to detect any significant differences in the results (Tukey's test, $p < 0.05$).

## Results and discussion

### Adsorption effect of residual feed on heavy metals

The residual feed in aquaculture wastewater is rich in substances such as crude fiber, starch, and protein, all of which have affinity for heavy metals (Hu et al. 2010). $Cu^{2+}$, $Cd^{2+}$ and $Pb^{2+}$ were selected as representative heavy metals in this experiment. Copper sulfate is a commonly-used disinfectant in the aquatic industry. According to the data from the China Ecological Environment Status Bulletin (Ministry of Ecology and Environment of the People's Republic of China 2023), the discharge of heavy metals Cd and Pb into natural waters is relatively large. By researching the adsorption of three typical heavy metal ions, $Cu^{2+}$, $Cd^{2+}$ and $Pb^{2+}$, the adsorption performance of residual feed on heavy metals in water was analyzed, which is the fundamental research for exploring the impact of heavy metals on the microbial degradation of residual feed.

#### *Adsorption of heavy metals by FAE and NFAE*

Before the adsorption experiment, there was insoluble precipitate at the bottom of the tube containing FAE wastewater, while there was no insoluble precipitate at the bottom of the tube containing NFAE wastewater. After the adsorption experiment, a large amount of sediment was retained at the bottom of FAE wastewater (Figure S1C), and a small amount of sediment was retained at the bottom of NFAE wastewater (Figure S1D). The adsorption efficiency of FAE wastewater on $Cu^{2+}$, $Cd^{2+}$, and $Pb^{2+}$were 61.64%, 70.14%, and 89.16%, respectively. The adsorption efficiency of NFAE wastewater on $Cu^{2+}$, $Cd^{2+}$, $Pb^{2+}$ were 4.29%, 2.98%, and 34.55%, respectively. It could be seen that the soluble components such as soluble protein and soluble starch in the residual feed also had the adsorption effect on heavy metals. Mejáre and Bülow (2001) also reported that various functional groups of proteins such as hydroxyl, amino, and thiol groups could form complex binding with heavy metal ions and proteins showed certain adsorption effect on heavy metals. However, the adsorption capacity of the soluble component was relatively small compared to the insoluble component. The insoluble components in the feed were crude fiber (mainly including cellulose, hemicellulose, lignin, and keratin), insoluble ash, and ether extract, etc. In other research, Chen et al. (2020) demonstrated that cellulose and hemicellulose had abundant hydroxyl functional groups, which could form coordination bonds with heavy metal ions and heavy metal ions could be adsorbed onto the surface of cellulose through interactions with cellulose, such as electro adsorption, complexation, and ion exchange. Rahman et al. (2020) treated electroplating wastewater containing heavy metals with cellulose materials, which could adsorb heavy metal ions from the solution and reduce heavy metal pollution in the environment and the removal efficiency reached 95%.

The adsorption ability of materials to different heavy metal ions often varies (Chai et al. 2021). The adsorption efficiency of FAE wastewater on three types of heavy metals was different. The adsorption efficiency on $Pb^{2+}$ was the strongest, while that on $Cu^{2+}$ was the weakest. On the other hand, the adsorption efficiency of NFAE wastewater on three types of heavy metals was also different. The adsorption efficiency on $Pb^{2+}$ was still the strongest, while that on $Cd^{2+}$ was the weakest. Thus, the residual feed has the greatest adsorption effect on heavy metal $Pb^{2+}$. Pb has a certain degree of biological toxicity, which can lead to the inhibition of microorganism growth, disruption of metabolism, damage to cell membranes, and obstruction of genetic material synthesis (Kasemodel et al. 2019). It could be seen that the adsorption of $Pb^{2+}$ by residual feed might have an impact on subsequent feed bio-degradation and ecological health, so further analysis was needed.

#### *Adsorption of heavy metals under different conditions*

The pH value affects the surface charge of materials, the chemical form and solubility of heavy metal ions in solution (Kicińska et al. 2022). Therefore, pH value may affect the adsorption of heavy metals by feed. Within the tested pH range (4–8), the adsorption capacity of residual feed on $Cu^{2+}$, $Cd^{2+}$, and $Pb^{2+}$ was not significantly affected by pH, so this result was not represented by a graph. Most natural water is neutral or weakly alkaline (Qiu et al. 2020), indicating that in natural waters, residual feed can have significant adsorption properties for three kinds of heavy metals. In addition, adsorption experiments were conducted under different temperatures. The results also showed that within the experimental temperature range, temperature had little effect on the adsorption of heavy metal ions by residual feed and this result was not represented by a graph. The effect of salinity on the adsorption effect of residual feed on heavy metals is illustrated in Fig. 1. As can be seen from the figure, when the salinity of the water increased, the adsorption capacity of residual feed on $Cu^{2+}$ and $Cd^{2+}$ decreased. An increase in salinity may lead to the competition between other ions in

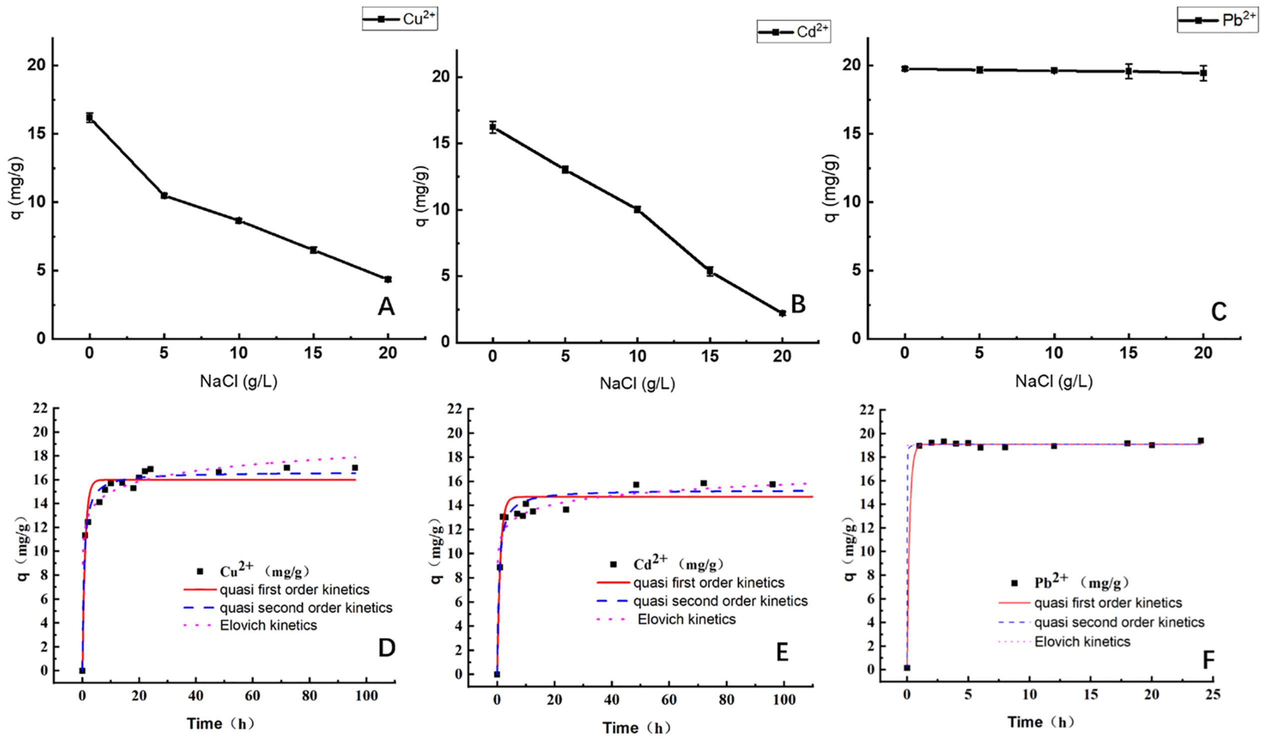



**Fig. 1** The adsorption effect of feed on heavy metals. (**A**: the effect of different salinities on the adsorption of $Cu^{2+}$; **B**: the effect of different salinities on the adsorption of $Cd^{2+}$; **C**: the effect of different salinities on the adsorption of $Pb^{2+}$; **D**: adsorption kinetic curve of $Cu^{2+}$; **E**: adsorption kinetic curve of $Cd^{2+}$; **F**: adsorption kinetic curve of $Pb^{2+}$)

the solution and heavy metal ions for adsorption on the material surface, which may reduce the effective sites available for heavy metal ion adsorption on the material surface (Barus et al. 2021). Also, the adsorption capacity would be reduced. However, compared with other heavy metals, the adsorption of $Pb^{2+}$ by residual feed was almost unaffected by salinity, and its adsorption capacity remained within the range of 19–20 mg/g under the experimental salinity. It can be seen that the feed had a strong binding ability with $Pb^{2+}$ and may not be affected by other ions, which is the same as the conclusion in Sect. "Adsorption of heavy metals by FAE and NFAE". However, in both fresh water and saline water, residual feed can adsorb heavy metals, so there may be a problem of heavy metal enrichment in feed during both freshwater and marine aquaculture.

### *Kinetic model of heavy metal adsorption*

This research used quasi-first-order kinetics, quasi-second-order kinetics, and Elovich kinetics to fit the adsorption process. Figure 1 shows the kinetic adsorption curves of the three metal ions. As seen from the figure, the adsorption capacity of $Cu^{2+}$ and $Cd^{2+}$ by residual feed increased at the initial stage, and then reached an equilibrium state at 48 h. The quasi-second-order kinetics was more suitable for the adsorption of $Cu^{2+}$ and $Cd^{2+}$ by residual feed (${R_{Cu}}^2=0.9847$; ${R_{Cd}}^2=0.9605$). Quasi-second-order kinetic adsorption showed that the adsorption process of residual feed on $Cu^{2+}$ and $Cd^{2+}$ mainly includes two processes surface diffusion and adsorption (Qi et al. 2019). Before adsorption begin, heavy metal ions must firstly diffuse to the surface of the adsorbent in the solution, which was influenced by the diffusion rate. Therefore, in the actual process, reducing the water flow velocity and mass transfer performance of the waters might help reduce the adsorption capacity. At the same time, the adsorption capacity of $Pb^{2+}$ by residual feed increased at the initial stage, and then reached an equilibrium state at 2 h. The quasi-first-order kinetics was more suitable for the adsorption of $Pb^{2+}$ by residual feed ($R^2=0.9989$). In quasi-first-order kinetics, the adsorption rate was directly proportional to the concentration of the adsorbed substance (Zhang et al. 2018). If the concentration of heavy metals in water increased, the adsorption rate would accelerate.

In summary, the soluble protein and starch of the residual feed dissolved in the waters and only adsorbed little heavy metals, and then the adsorbed complex slowly settled at the bottom and accumulated with the FAE of the residual feed at the bottom. The insoluble fraction of residual feed had a significant adsorption effect on all three typical heavy metal ions. The adsorption for heavy metals was almost unaffected by environmental pH and temperature. However, as salinity increased, the adsorption effect on $Cu^{2+}$and $Cd^{2+}$ decreased, while the adsorption effect on $Pb^{2+}$ was not changed significantly. Residual feed could quickly adsorb heavy metals and reach adsorption saturation within 48 h. Because residual feed was not a conventional adsorbent, its adsorption capacity for heavy metals was lower than 20 mg/g. Compared to other adsorbent products, its adsorption capacity is not large. For example, Madivoli et al. (2016) reported that citric acid modified cellulose had the ability to adsorb and remove heavy metals and the adsorption capacity was 18.06 mg/g $Cu^{2+}$, 42.69 mg/g $Cd^{2+}$, and 21.64 mg/g $Pb^{2+}$ respectively. Repo et al. (2011) explored the adsorption potential of the synthesized modified chitosan material with $Co^{2+}$, $Ni^{2+}$, $Cd^{2+}$, and $Pb^{2+}$ as target metals. Under their experimental conditions, the maximum adsorption capacity of the obtained hybrid material for metal ions was 0.25–0.63 mmol/g. Ma et al. (2015) prepared porous starch xanthate (PSX) and porous starch citrate (PSC) adsorbents, and the adsorption behavior of PSX and PSC on lead ions was in accordance with the pseudo-second-order kinetic model. When the preparation conditions were optimized, the maximum adsorption capacities of PSX and PSC reached 109.1 mg/g and 57.6 mg/g, respectively. This research has confirmed the adsorption process of heavy metals by residual feed, especially the fast adsorption of $Pb^{2+}$ by residual feed. Moreover, the enrichment of heavy metals by residual feed may bring certain negative ecological effects, and further research is needed.

## The microbial degradation of residual feed

In aquaculture waters, residual feed, feces, and other metabolites at the bottom of the pond rely on the microorganisms in the waters for degradation. Due to the high content of FAE such as crude fiber and soluble substances such as soluble starch and protein in the feed, when the residual feed was discharged into natural waters with wastewater, the organic load of waters would increase. The previous research results (Sect. "Adsorption effect of residual feed on heavy metals") showed that FAE, soluble starch, and soluble protein in the residual feed could adsorb heavy metals, especially fibers and other insoluble components. The preliminary experiments have shown that heavy metal ions can have a certain negative impact on the growth and metabolism of *Bacillus licheniformis*. It remains unknown about whether this adsorption can affect the degradation efficiency of soluble starch and soluble protein in the residual feed by microorganisms. In response to this question, the experiments and analysis were conducted.

The preliminary research results of the research group showed that *Bacillus licheniformis* can efficiently degrade soluble proteins and starch in the residual feed. This research analyzed the impact of heavy metals on the degradation of residual feed by *Bacillus licheniformis*. The result is shown in Figs. 2, 3, 4. A significant difference can be observed in the effect of heavy metals on the degradation of residual feed by *Bacillus licheniformis* in wastewater containing insoluble contents (FAE) and wastewater removing insoluble contents (NFAE). Meanwhile, the results of microbial degradation should be analyzed in conjunction with the previous adsorption results to better assess the impact of adsorption on degradation.

### *Effect of $Cu^{2+}$ on microbial degradation of residual feed*

$Cu^{2+}$ had a certain antibacterial effect, and copper sulfate was also a common disinfectant. $Cu^{2+}$ can interfere with the integrity of microbial cell membranes and walls, affect cell function, and inhibit microbial growth and reproduction. However, the concentration of $Cu^{2+}$, as well as the types of microorganisms and environmental conditions, can also affect the degree of impact (Ochoa-Herrera et al. 2011). As shown in Fig. 2, the presence of $Cu^{2+}$ in both NFAE and FAE wastewater had a certain inhibitory effect on the growth and degradation of microorganisms. However, for the microbial degradation process, the inhibitory effect of $Cu^{2+}$ in the NAFE system was significantly greater than that in the FAE system. According to the previous adsorption experiment, it was known that the insoluble substances in the FAE system had a strong adsorption effect on $Cu^{2+}$. After being adsorbed, the $Cu^{2+}$ settled to the bottom, reducing the impact of $Cu^{2+}$ on microorganisms in the solution. However, because $Cu^{2+}$ was adsorbed by the insoluble component in the residual feed, bacterial biomass would be less affected by $Cu^{2+}$. Besides, the biomass of *Bacillus* should remain at a high level in the high concentration $Cu^{2+}$-FAE wastewater system. But this speculation was very different from the actual results. The experimental results showed that the biomass of *Bacillus licheniformis* in wastewater with $Cu^{2+}$ at a concentration of 50.0 mg/L after 7 days was much lower than the biomass of *Bacillus subtilis* in FAE wastewater without $Cu^{2+}$. It could be inferred that some bacterial cells might also be adsorbed by the insoluble component and then settled to the bottom of the water. So, the sediment adsorption group at the bottom of the water should be the FAE adsorbing $Cu^{2+}$ and the bacterial cell (B) adsorption complex FAE-B-$Cu^{2+}$. The adsorption process between microbial cells and heavy metal ions was also reported in other research (Barik et al. 2021), so this speculation should be reasonable.

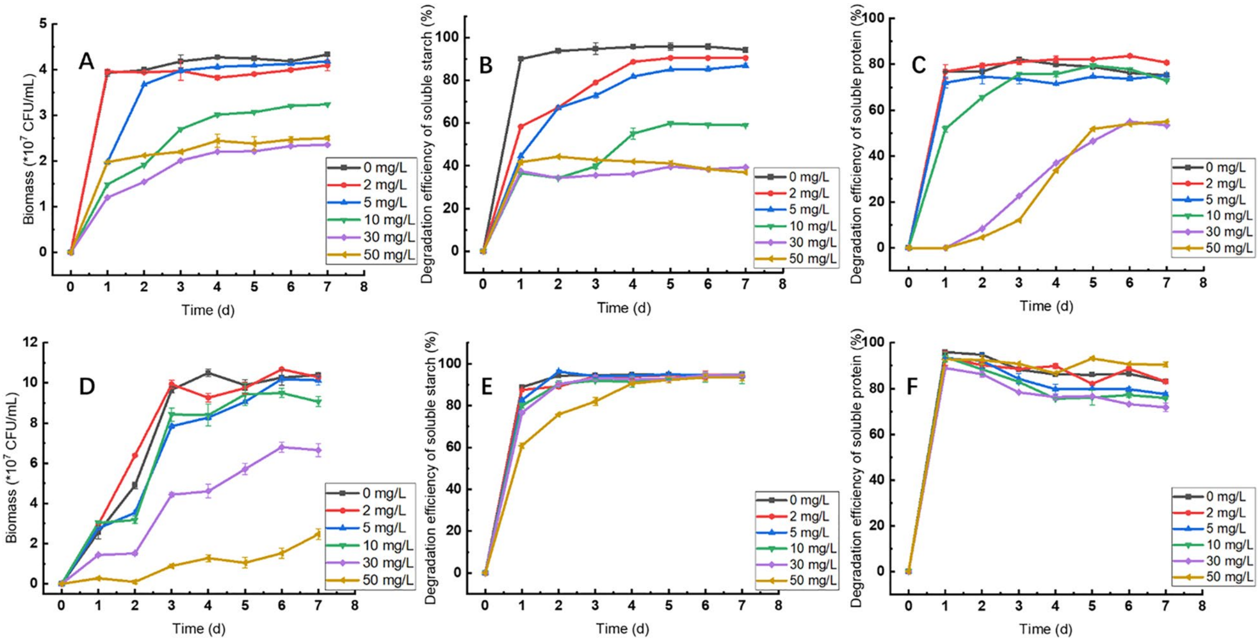



**Fig. 2** The effect of $Cu^{2+}$ on microbial degradation of residual feed. (**A**: microbial biomass in the NFAE system; **B**: soluble starch degradation in the NFAE system; **C**: soluble protein degradation in the NFAE system; **D**: microbial biomass in the FAE system; **E**: soluble starch degradation in the FAE system; **F**: soluble protein degradation in the FAE system)

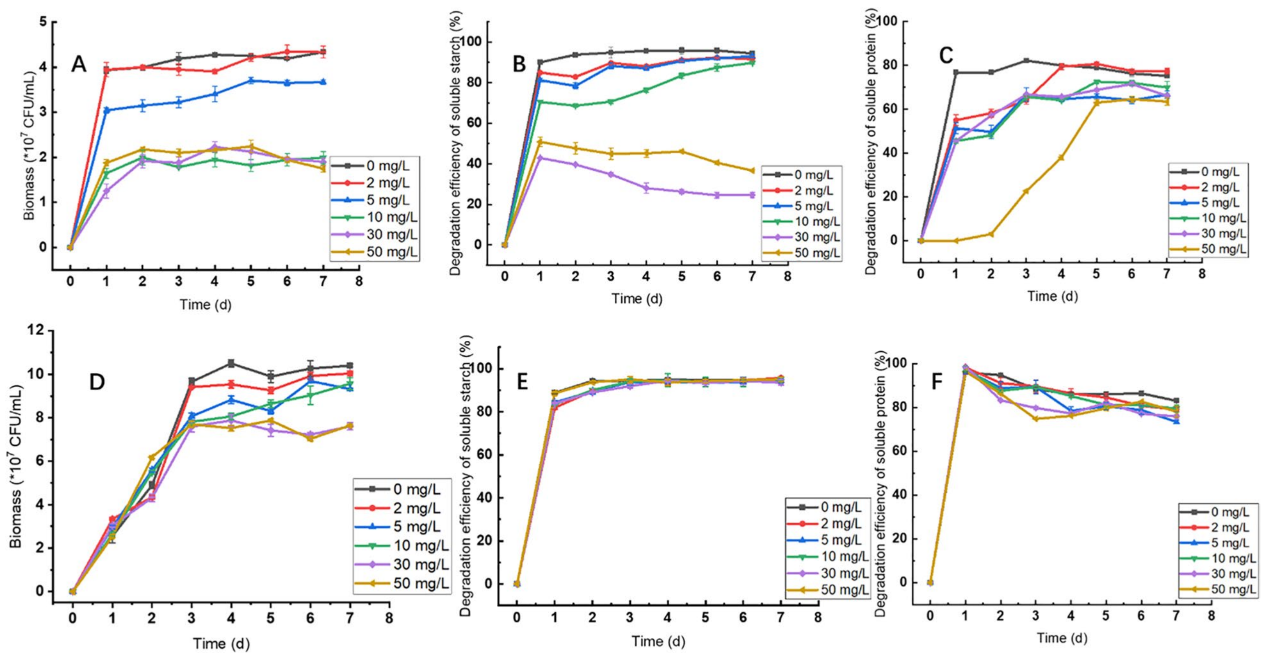



**Fig. 3** The effect of $Cd^{2+}$ on microbial degradation of residual feed. (**A**: microbial biomass in the NFAE system; **B**: soluble starch degradation in the NFAE system; **C**: soluble protein degradation in the NFAE system; **D**: microbial biomass in the FAE system; **E**: soluble starch degradation in the FAE system; **F**: soluble protein degradation in the FAE system)

In addition, there was a strange phenomenon of *Bacillus licheniformis* degrading soluble proteins in FAE wastewater. With the higher concentration of $Cu^{2+}$ and the lower microbial biomass, the degradation efficiency of soluble protein and starch was still high on the 7th day. Based on the experimental results in Sect. "Kinetic model

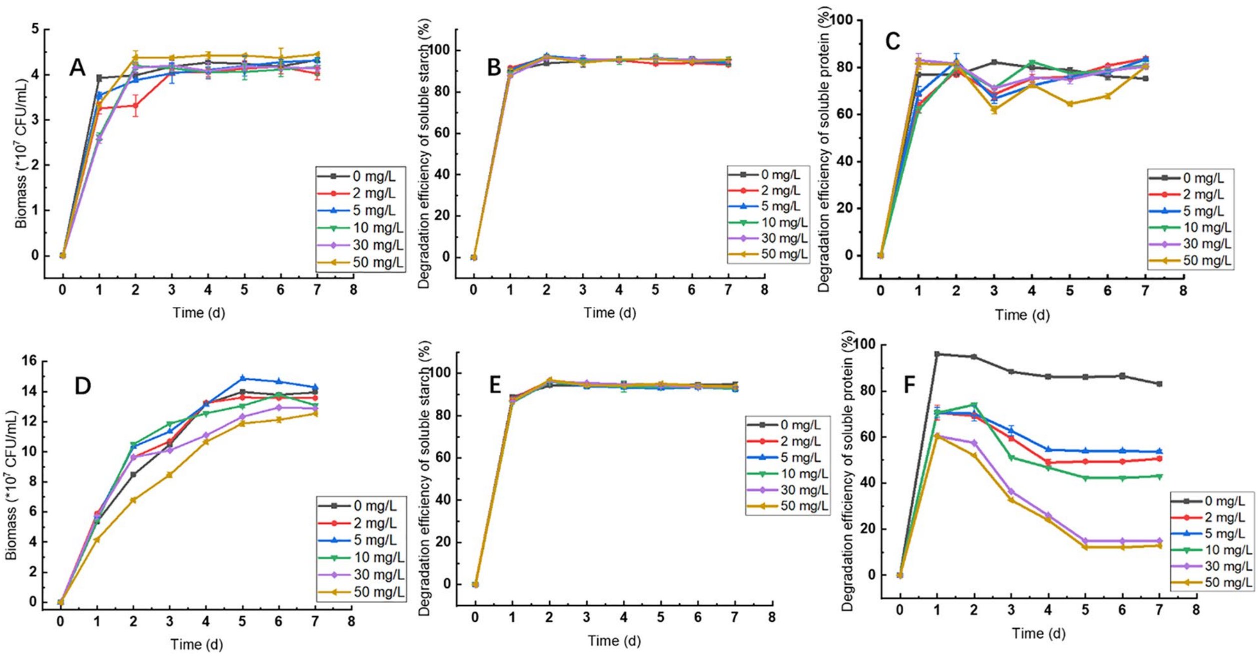



**Fig. 4** The effect of $Pb^{2+}$ on microbial degradation of residual feed. (**A**: microbial biomass in the NFAE system; **B**: soluble starch degradation in the NFAE system; **C**: soluble protein degradation in the NFAE system; **D**: microbial biomass in the FAE system; **E**: soluble starch degradation in the FAE system; **F**: soluble protein degradation in the FAE system)

of heavy metal adsorption", it was deduced that $Cu^{2+}$ could also be adsorbed by soluble components, and the complex settled to the bottom of the cup and was isolated from the wastewater.

It could be seen that the composition of the adsorption complex in this system not only included insoluble components (FAE) and heavy metals (M), but also includes bacteria (B), soluble components (proteins and starch). The adsorption complex (FAE-B-S-Pr-$Cu^{2+}$) would be settled to the bottom. If in natural waters, the adsorption complexes might become a part of the sediment. The subsequent scanning electron microscopy and energy spectrum analysis was carried out to prove this point.

### *Effect of $Cd^{2+}$ on microbial degradation of residual feed*

$Cd^{2+}$ can compete with other ions on microbial cell membranes, blocking the absorption and utilization of other elements such as calcium and magnesium (Vig et al. 2003). This can lead to an imbalance of elements within microbial cells, affecting their growth and metabolism. High concentrations of $Cd^{2+}$ can also disrupt the structure and function of microbial cells, leading to cell death (Khan et al. 2022). As shown in Fig. 3, the presence of $Cd^{2+}$ in both NFAE and FAE wastewater had a certain inhibitory effect on the growth and degradation of microorganisms. The effect of $Cd^{2+}$ on the degradation of residual feed by *Bacillus licheniformis* was basically consistent with that of $Cu^{2+}$.

### *Effect of $Pb^{2+}$ on microbial degradation of residual feed*

The adsorption ability of materials to different metals depends on various factors, including their chemical properties, structural characteristics, and the physical properties of metals (Chai et al. 2021). In Sect. "Adsorption effect of residual feed on heavy metals", it was confirmed that the feed had a strong adsorption capacity for $Pb^{2+}$. The adsorption of $Pb^{2+}$ in FAE wastewater and NFAE wastewater was different from $Cu^{2+}$ and $Cd^{2+}$, so the effect of $Pb^{2+}$ on microbial degradation of residual feed was different compared to other heavy metals (Fig. 4).

In NFAE wastewater, the degradation of soluble starch was not affected by the concentration of $Pb^{2+}$. After the 7 days of degradation, the degradation efficiency of soluble starch by *Bacillus licheniformis* at all concentrations of $Pb^{2+}$ was ranged from 93.09 to 95.68%, which was basically consistent with the experimental results of the blank control group. This result was significantly different from other heavy metal ions. In addition, the degradation efficiency of soluble protein was also little affected by $Pb^{2+}$. From this, it can be inferred that a large amount of $Pb^{2+}$ in NFAE wastewater was adsorbed by soluble components in the residual feed, forming adsorption groups that settled at the bottom of the water. The $Pb^{2+}$ isolated from the water and the negative impact on microorganisms was reduced. This result was also consistent with the adsorption experimental results of heavy metals in

Sect. "Adsorption of heavy metals by FAE and NFAE". From the adsorption experimental results, it can be known that both the insoluble and soluble components in the residual feed had the strongest adsorption ability for $Pb^{2+}$ among the three heavy metal ions. In addition, due to the soluble components of residual feed in NFAE wastewater adsorbing $Pb^{2+}$, forming adsorption complex that settle at the bottom of the water, the negative impact of $Pb^{2+}$ on microorganisms was reduced, therefore the biomass of *Bacillus subtilis* was not significantly affected by changes in $Pb^{2+}$ concentration.

The degradation of residual feed by *Bacillus licheniformis* by $Pb^{2+}$ in FAE wastewater was similar to other heavy metal ions. From the changes in the degradation efficiency of soluble starch and protein in FAE wastewater and NFAE wastewater, it could be seen that the soluble component that adsorbed $Pb^{2+}$ should be mainly soluble starch but not protein, because the overall removal efficiency of soluble protein in FAE wastewater was not high (12.85–53.65%) at 7th day. However, the removal of soluble starch in FAE wastewater and NFAE wastewater was not different from the blank experiment. In the research by Gunawardene et al. (2021), it was also confirmed that starch-based materials had a good adsorption effect on $Pb^{2+}$ and the maximum adsorption capacity of two modified cassava starch samples for $Pb^{2+}$ could reach 370.37 mg/g and 294.12 mg/g, respectively.

### SEM–EDS analysis of adsorption complex

The SEM–EDS results of the adsorption complex inferred in Sect. "The microbial degradation of residual feed" are shown in Figs. 5 and 6. The surface morphology of the feed was relatively smooth, and the energy spectrum showed that the composition elements of the feed were mainly C, O, N, Si and P, which were consistent with the composition. The SEM images and energy spectrum analysis of the feed-*Bacillus licheniformis* showed that a large number of bacterial cells were adsorbed by the residual feed, and the energy spectrum showed that its elemental composition was mainly C, O, N and P. In the microbial degradation system containing heavy metals, not only bacterial cells were adsorbed by the feed, but also heavy metals were adsorbed by the feed, forming a complex of heavy metals, feed components, and microbial cells. Energy spectrum analysis also confirmed the adsorption process of metals (Fig. 6). In other studies, it was also reported that heavy metal elements could combine with organic matter and microbial cells to form complex matters. For example, Violante et al. (2010) reported the adsorption/desorption reactions of biotic and abiotic organisms in soil environments, as well as the chemical complexation processes with inorganic and organic ligands, indicating that the interaction between chemical and biological interfaces had a certain impact on the bioavailability and mobility of heavy metals in soil. Yang et al. (2023) studied the heavy metal treatment process in artificial wetlands and proposed that the adsorption/surface complexation and binding/assimilation of microorganisms and inorganic and organic components played a dominant role in the metal removal potential of wetlands.

According to previous speculations, $Pb^{2+}$ was more easily adsorbed by starch, while $Cu^{2+}$ can be adsorbed by protein and starch. From the EDS energy level stratification diagram (Fig. 5), it can be seen that the density of

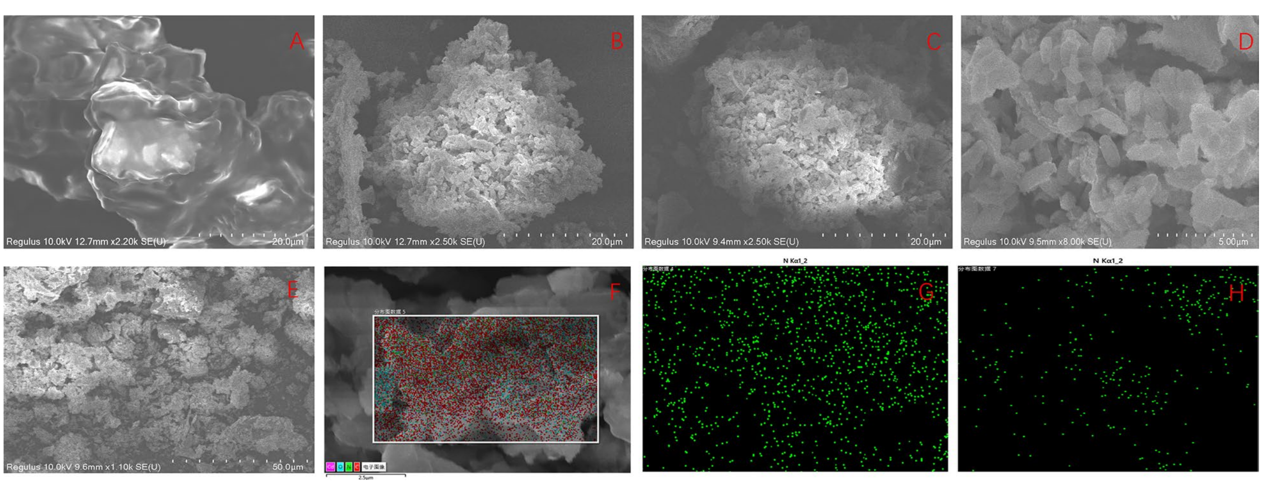



**Fig. 5** Scanning electron microscope-Energy dispersive spectrometer of feed and adsorption complex (**A**: SEM image of feed; **B**: SEM image of feed-*Bacillus licheniformis*; **C**: SEM image of feed-*Bacillus licheniformis*-Cu; **D**: SEM image of feed-*Bacillus licheniformis*-Cd; **E**: SEM image of feed-*Bacillus licheniformis*-Pb; **F**: EDS image of feed-*Bacillus licheniformis*-Cd; **G**: EDS image of N distribution in feed-*Bacillus licheniformis*-Cu system; **H**: EDS image of N distribution in feed-*Bacillus licheniformis*-Pb system)

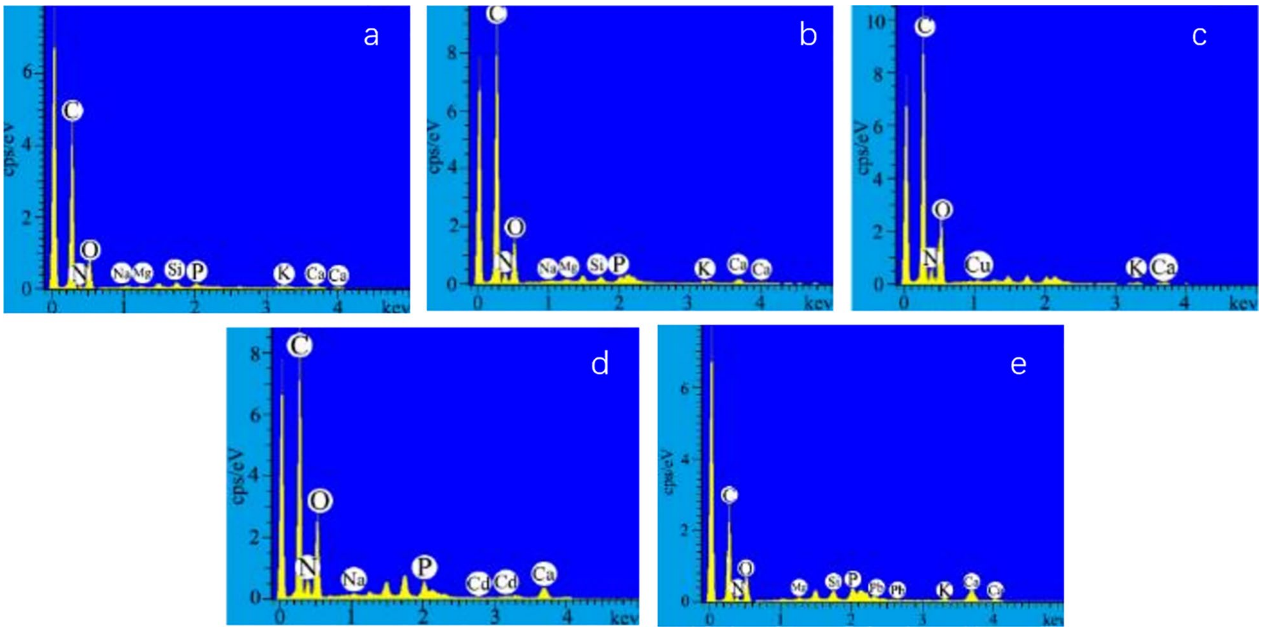



**Fig. 6** EDS images of feed and adsorption complex (**a**: feed; **b**: feed-*Bacillus licheniformis*; **c**: feed-*Bacillus licheniformis*-Cu; **d**: feed-*Bacillus licheniformis*-Cd; **e**: feed-*Bacillus licheniformis*-Pb)

N in the $Cu^{2+}$ adsorption system was much higher than $Pb^{2+}$ adsorption system, which can once again verify the hypothesis.

Overall, both the insoluble and easily soluble components in the residual feed could adsorb heavy metals and bacterial cells, forming FAE-B-S-Pr-metal adsorption complex. As the adsorption groups settle, some soluble starch and protein could also be isolated from the water and settled at the bottom. The adsorption effect could reduce the negative impact of heavy metals in the water on microbial degradation of residual feed. However, in-depth research was needed to determine whether heavy metals present in sediment would interfere with the degradation of pollutants by sediment microorganisms, thereby affecting the structure of benthic communities and causing negative environmental impacts.

#### Microbial degradation of residual feed in natural waters

The existence of heavy metals in the environment might affect the metabolic activity of environmental microorganisms in degrading organic matters. To simulate the actual situation, this research explored the impact of heavy metals on the microbial degradation of residual feed in natural waters (freshwater and seawater) based on the research in Sect. "The microbial degradation of residual feed" (Fig. 7).

As shown in Fig. 7, the presence of heavy metals had a certain negative impact on the microbial degradation of residual feed, and the impact was greater in the seawater environment. According to the conclusion in Sect. "Adsorption of heavy metals under different conditions", the increase in salinity would weaken the adsorption of heavy metals by the feed. Therefore, in the seawater environment, the adsorption and sedimentation between heavy metals and the feed were weaker, which led to the greater impact of heavy metals on the microbial degradation of organic matters. Besides, unlike the degradation of soluble starch, the degradation efficiency of soluble protein decreased with the extension of time, indicating that there might be a continuous release of protein during the treatment process. The above experimental results verified the adsorption effect of residual feed on heavy metals again, and the adsorption complex would settle at the bottom of the waters. This was consistent with the phenomenon of heavy metals accumulating in natural sediment (Hang et al. 2009). From this research, it could be seen that the aquaculture feed was discharged into the surrounding water environment with the aquaculture water, and the heavy metals in the water environment could be adsorbed by the feed and accumulated in the sediment.

In natural waters (rivers, lakes, and seas), especially during the wet season, the rapid flow velocity may affect the sedimentation of the adsorption complex and the complex would be in a suspended state. In the case, the heavy metal-enriched substance was highly likely to be consumed by large aquatic animals and then enter the food chain (Li et al. 2022). In addition, heavy metals

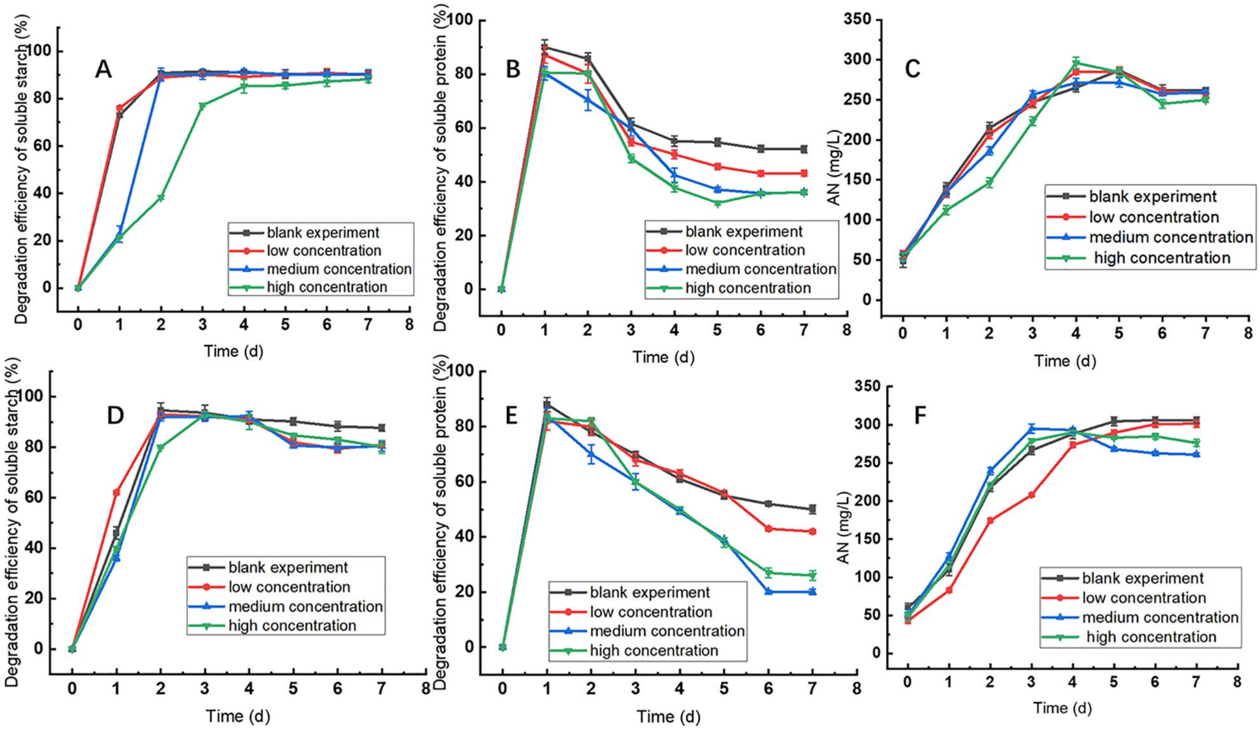



**Fig. 7** The effect of heavy metals on microbial degradation of residual feed in natural waters (**A**: soluble starch degradation in freshwater system; **B**: soluble protein degradation in freshwater system; **C**: AN of freshwater system; **D**: soluble starch degradation in seawater system; **E**: soluble protein degradation in seawater system; **F**: AN of seawater system)

can affect the biodegradation of pollutants in sediment, thereby affecting the community structure of benthic organisms. Complex substances containing heavy metals may also be consumed by benthic organisms and enter economic aquatic organisms along the food chain, causing negative impacts on human health. Heavy metals in sediment may also be released into waters, causing contamination again (Liu et al. 2019).

Therefore, the collection and centralized treatment of aquaculture residues deserve more attention. Although the environmental hydrological conditions such as water flow and wind waves in natural waters (rivers, lakes, and seas) are different from experimental environments, the results of this experimental research contribute to the standardized management of residual feed wastewater.

## Conclusion

The feed is possible to adsorb heavy metals in water and the degradation of residual feed and the restoration of the water environment would be affected. The experimental results of the adsorption effect of residual feed showed that both insoluble component (FAE) and soluble component (soluble protein and starch) had adsorption effects on heavy metals. Under experimental conditions, there was no significant difference in the adsorption of heavy metals by residual feed under different environmental pH and temperature. However, as the salinity increased, the adsorption capacity of $Cu^{2+}$ and $Cd^{2+}$ decreased, while the adsorption effect on $Pb^{2+}$ was not changed significantly. Both the insoluble and easily soluble components in the residual feed could adsorb heavy metals and bacterial cells, forming FAE-B-S-Pr-metal adsorption complex. The adsorption effect could reduce the heavy metal load in the water body and reduce the negative impact of heavy metals on microbial degradation of residual feed in the water body, but heavy metals would accumulate in the sediment of residual feed. Further research is needed to investigate the interference caused by the adsorption of heavy metals by residual feed on the degradation of pollutants by sediment microorganisms, as well as its negative effects on the community structure of benthic organisms and environment.

## Supplementary Information

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Supplementary Material 1.

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Not applicable.

**Author contributions**
Sufeng Wang and Wen Zhang conceptualized of the study, participated in its design and coordination, and drafted the manuscript. Zijing Wang carried out most of the tests. Jingshan Li and Mingyang Lin completed some supplementary experiments. Li Chen gave some assistance to the experiments. All authors read and approved the final manuscript.

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**Availability of data and materials**
The data are available from the corresponding author on reasonable request.

## Declarations

**Ethics approval and consent to participate**
Not applicable.

**Consent for publication**
All authors approved the final manuscript and the submission to this journal.

**Competing interests**
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.

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