Effect of Vegetable Growth on Content and Composition of Antibiotics in *Litopenaeus vannamei* Pond Sediments in Crop/Aquacultural Rotation Process

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Abstract: Photodegradation remains the major pathway of antibiotic removal in natural ponds. This study introduced a new method of growing vegetables on the bottom substrate of shrimp ponds to improve sediment quality. The aim of the study was to investigate the effect of vegetable planting on the photodegradation of antibiotics. This study characterized antibiotic levels in the pond sediment during this phytoremediation process and investigated the antibiotic content and composition of the sediment with and without crop rotation (traditional control), as well as the shrimp yields. The results showed that total antibiotics (e.g., trimethoprim, oxytetracycline, and norfloxacin) in the sediment of all aquaculture ponds continuously decreased from 44.78 ± 4.07 µg/kg to 18.80 ± 2.26 µg/kg in the crop rotation pond. The total amount of antibiotics consistently decreased in all ponds, and the rate of decline did not greatly differ. However, oxytetracycline in the crop rotation pond decreased faster than in the control pond, presumably because the growing vegetables altered the sediment and microbial-community characteristics that promoted oxytetracycline degradation. In the following year, there was little difference in the levels of norfloxacin or oxytetracycline between the two ponds. An increase in trimethoprim in the control pond was much higher than in the crop-growing sediment. It was indicated that the system remediated the shrimp pond ecosystem as well as providing the possibility of increasing profits by planting vegetables in the winter idle period of shrimp ponds.

Keywords: *Litopenaeus vannamei*; aquaculture pond; sediment; antibiotic; in situ improved

1. Introduction

As the world’s demand for aquatic products increases, there is great pressure on the environment from the use of such chemicals as insecticides and biocides [1,2]. In addition, heavy metal pollution, eutrophication, and harmful algal blooms (HABs) are other common environmental challenges associated with aquaculture [3–6]. Coastal aquaculture (e.g., near China’s Hangzhou Bay) in particular is a challenge. Shrimp disease outbreaks occur frequently and trigger the excessive use of antibiotics that further compromises the quality of the aquatic ecosystem as well as human health [7–9]. Diverse antibiotics are added to water to fight against pathogenic microbes during breeding [10,11], and those remaining or excessive (more than 30%) antibiotics thus accumulate in the sediment and bioaccumulate in fish, shrimp, and aquaphytes [12–15]. For instance, erythromycin was found to vary from 0.8 ng/g to 4.8 ng/g in sediment samples from aquaculture areas in Hailing Island in Southern China [9]. Oxytetracycline (OTC) and enrofloxacin (ENR) in sediment in the Tha Chin and Mun rivers have concentrations ranging from 4.5 µg/kg to 4062 µg/kg and from 1.4 µg/kg to 2239 µg/kg, respectively [16]. Wang et al. detected nine antibiotics in the aquacultural feed, including sulfamethoxazole (SMX), levofloxacin (LEOF), oxytetracycline (OTC), chlortetracycline (TC), norfloxacin (NOF), and tetracycline (TC) with concentrations ranging from 1.10 µg/kg to 46.45 µg/kg.
Pond sediment remediation is essential in restoring ecological functions. The most common practices are clearing ponds, sunlight exposure, and dredging excessive sediment [17]. However, these methods suffer from the problems of sediment disposal and limited antibiotic photodegradation after mixing with sediment [18,19]. Although sediment microbes can degrade antibiotics, the biodegradation process is limited by temperature and kinetics [20]. Photodegradation remains the major pathway of antibiotic removal in natural ponds [21,22]. Recently, a shrimp–vegetable rotation or alternation cultivation method was tested in which the shrimp culture ponds were used to grow vegetables (e.g., broccoli, potherb mustard, lettuce, and Lactuca saliva) in winter, as the root systems absorb and enrich antibiotics through the passive-diffusion effect [23]. Ni assessed the economic and ecologic efficiency of the new shrimp–vegetable rotational farming system model. It was found that the rotational farming system significantly reduces the TN and TP contents and phytoplankton community in soil and water and achieves a considerably better economic performance compared with that of traditional monoculture farming. Specifically, citric acid or glycine from plant root exudates were reported to play an important role in the utilization and degradation of exogenous antibiotics by changing microbial populations and structures around the plant rhizosphere [24,25]. However, it was unclear whether the shading of vegetable growth would affect the photodegradation of antibiotics in these ponds.

This study characterized the antibiotic residues in pond sediment with and without the planting of *Litopenaeus vannamei*, which was selected because of its short growth time, low investment, and high yield. We measured the total levels and compositions of target antibiotics at different time stages and explored reasons for their reduction and the improvement of pond sediment quality through vegetable cultivation.

2. Materials and Methods

2.1. Collection, Preservation, and Pretreatment of Sediment Samples

The aquaculture ponds (121°5′10″, 30°17′34″) studied in this project are located on the south bank of Hangzhou Bay, China. Broccoli (*Brassica oleracea*) and potherb mustard (*Brassica juncea*) were grown in the pond sediment over the winter (from December 2017 to April 2018) and were selected due to their short growth time, low investment, and high yield. Three ponds were selected as the control group with traditional pond maintenance (ploughing and drying), and six ponds were experimental sites treated with shrimp–vegetable rotational farming (three ponds for broccoli and three ponds for potherb mustard) as shown in Figure 1. The broccoli and potherb mustard planting density in the experimental ponds comprised 62,500 plants/hm² and 83,250 plants/hm², respectively. Three parallel samples were collected in each pond in December 2017, March 2018, April 2018, and August 2018, which corresponded to the four phases of vegetable planting, the mid-stage of the plant growth, post-plant harvest, and the end of shrimp farming, respectively. Sediment sampling was performed at five different pond locations using a mud picker at the plowing height of 20–30 cm with a pond water depth of typically 0.7–1.1 m. No additional fertilizers or pesticides were used during the vegetable planting. The sediment samples (approximately 1 kg) were stored in polyethylene bags and freeze-dried for 36 h, and the solid samples were ground and sieved (100 mesh size). The powder samples were saved in blue-cap glass bottles and stored at −20 °C for further extraction.

2.2. Extraction and Detection Methods for Antibiotics

Based on relevant reports on antibiotic residue in sediments in Hangzhou Bay [26], this study selected 12 typical antibiotics for detection: sulfadiazine (SDZ), sulfamethoxine (SMD), sulfamethazine (SMZ), sulfamethoxazole (SMX), norfloxacin (NOF), tetracycline (TC), oxytetracycline (OTC), chlortetracycline (CTC), erythromycin (ETM), roxithromycin (RTM), trimethoprim (TMP), and levofloxacin (LEOF). The standards of TMP and 13C-TMP purchased from Sigma-Aldrich (USA) were used as recovery indicators. The LEOF standard was purchased from Tokyo Chemical Industry (TCI), while the other ten antibiotics were
purchased from Dr. Ehrenstorfer (Germany). All standard products were solid powder with purity ≥ 97%.

![Figure 1. Layout of aquaculture ponds in the study area.](image)

The pretreatment and extraction were conducted as follows: Each sediment sample of 4.00 g was accurately weighed and placed in a centrifuge tube, and 10 mL of the methanol/citric acid buffer (1:1, v/v) extraction solution was added to the tube. The samples were mixed in a vortex mixer for 1 min and then ultrasonically extracted for 15 min followed by centrifugation (4000 r/min for 5 min). The supernatant was transferred to a flat-bottomed flask using a long glass Pasteur pipette. This operation was repeated three times to increase the antibiotic extraction. The supernatant was evaporated using a rotary evaporator to a constant volume of 49 µL.

After the solid-phase extraction, samples were concentrated by nitrogen blowing the elute until the volume was reduced to 10%. The elute was then adjusted to 1 mL with methanol, filtered (syringe filter 0.22 µm, organic phase, Hengqi Company, Shanghai, China), and stored in a refrigerator at 4 °C prior to the high-performance liquid chromatography mass spectrometry (HPLC-MS/MS) analysis. An Agilent ZORBAX Eclipse XDB-C18 (150 mm × 2.1 mm, 5 m) was used with a column temperature of 30 °C. The A and B mobile phases were methanol and 0.1% acetic acid aqueous solution, respectively, and applied as shown in Table 1. The injection volume was 10 µL at a flow rate of 0.4 mL/min.

| Time (min) | A (%) | B (%) |
|-----------|-------|-------|
| 0         | 30    | 70    |
| 2.0       | 60    | 40    |
| 4.0       | 90    | 10    |
| 8.0       | 90    | 10    |
| 8.5       | 30    | 70    |
| 12.0      | 30    | 70    |

Table 1. Sequence of mobile phase gradients in HPLC-MS/MS.
The positive-ion scanning mode of multiple reaction monitoring (MRM) was used in mass spectrometry. The temperature of the transmission capillary was 350 °C, and the voltage of capillary was 4500 V. The flow rates of sheath gas (N₂) and auxiliary gas (N₂) were 30 arb and 10 arb, respectively, and the atomizer pressure was 40 psi. The ion monitoring conditions of the different antibiotics are shown in Table 2.

Table 2. Ion monitoring conditions of antibiotics.

| Antibiotics | Parent Ion (m/z) | Daughter Ion (m/z) | Retention Time (m/z) | Fragmentor Voltage (V) | Collision Energy (eV) |
|-------------|-----------------|-------------------|---------------------|------------------------|----------------------|
| SDZ         | 250.9           | 108.0, 155.9      | 1.26                | 72                     | 31, 17               |
| SMX         | 253.8           | 92.1, 108.0       | 2.44                | 65                     | 26, 16               |
| SMZ         | 278.9           | 124.0, 186.0      | 2.08                | 72                     | 26, 17               |
| SMD         | 280.9           | 92.1, 155.9       | 1.89                | 76                     | 28, 17               |
| TMP         | 291.0           | 230.8, 261.8      | 1.33                | 100                    | 24, 25               |
| 13C-TMP     | 294.2           | 231.8, 262.8      | 1.35                | 107                    | 23, 24               |
| NOF         | 319.4           | 275.9, 301.8      | 1.77                | 113                    | 15, 20               |
| LEOF        | 362.2           | 261.2, 317.9      | 1.60                | 114                    | 27, 18               |
| TC          | 442.8           | 340.8, 414.7      | 4.52                | 132                    | 48, 31               |
| OTC         | 461.3           | 222.9, 426.6      | 9.69                | 113                    | 58, 19               |
| CTC         | 478.3           | 443.6, 461.6      | 3.12                | 109                    | 18, 17               |
| ETM         | 716.0           | 158.0, 558.4      | 4.47                | 109                    | 30, 16               |
| RTM         | 837.8           | 157.8, 679.0      | 4.48                | 131                    | 32, 19               |

2.3. Quality Control and Quality Assurance

A series of standard solutions with concentrations of 1, 2, 5, 10, 20, 50, and 100 µg/L were used to optimize the LC/MS operational conditions. The target compounds were quantified by calibration curves using the external standard method in which the peak area and the concentration of each component (µg/L) were set as the X and Y axis, respectively. The linear correlation coefficients between the concentration and the peak area were all higher than 0.99 (Table 3).

Table 3. Regression equations, correlation coefficients, and detection limits of the tested antibiotics.

| Antibiotic | Concentration–Peak Area Linear Equations | \( R^2 \) | Detection Limit |
|------------|------------------------------------------|----------|----------------|
| SDZ        | \( y = 0.00005773x - 0.4445 \)           | 0.9991   | 0.017          |
| SMX        | \( y = 0.00006452x - 0.6231 \)           | 0.9992   | 0.015          |
| SMZ        | \( y = 0.00004717x - 1.214 \)            | 0.9996   | 0.014          |
| SMD        | \( y = 0.00006818x - 0.8433 \)           | 0.9992   | 0.028          |
| TMP        | \( y = 0.00350072x - 0.5668 \)           | 0.9990   | 0.009          |
| 13C-TMP    | \( y = 0.00008375x - 0.3072 \)           | 0.9987   | 0.009          |
| NOF        | \( y = 0.00032399x + 1.272 \)            | 0.9997   | 0.217          |
| LEOF       | \( y = 0.00004277x - 0.7590 \)           | 0.9986   | 0.033          |
| TC         | \( y = 0.00010161x + 0.3603 \)           | 0.9991   | 0.108          |
| OTC        | \( y = 0.00117097x - 2.3480 \)           | 0.9993   | 0.094          |
| CTC        | \( y = 0.00019284x + 0.1712 \)           | 0.9991   | 0.055          |
| ETM        | \( y = 0.00003306x - 0.5322 \)           | 0.9994   | 0.006          |
| RTM        | \( y = 0.00020047x - 1/1833 \)           | 0.9999   | 0.007          |

In the spiked and recovery experiments, we used quartz sand as the matrix in the blank sample to evaluate the precision and accuracy of the extraction and detection processes. Added to 4.00 g of quartz sand were 40 ng and 200 ng of the spiked antibiotics (40 µL and 200 µL from the mixed standard with 1 ppm target standards), respectively. The experiments were repeated 3 times for each spiked concentration. The results showed that the recovery rates of 12 antibiotics in the quartz sand matrix were 78.45%~112.99%, and the relative standard deviations were below 12%.
According to the International Conference on Harmonization of Technical Requirements for Registration of Pharmaceuticals for Human Use Q2 (ICHQ2) guidelines, the response intensity (peak height) of the sample of low concentrations (e.g., 10 ng/g) and the blank sample were used to calculate the detection limit given a signal ratio of 3:1. The response value of the blank sample was selected before and after the retention time. The detection limits of target antibiotics in the sediments were between 0.006 ng/g and 0.27 ng/g based on the measurement results of both the blank samples and the samples.

3. Results and Discussion
3.1. Concentration Changes of Sediment Antibiotics in the Shrimp–Vegetable Rotational Farming Period

Figure 2 shows the antibiotic concentrations in sediments of both the control and experimental ponds from December 2017 to April 2018. The antibiotic concentrations dropped continuously over time in all the ponds, with the average concentration decreasing from 44.78 µg/kg (39.98 µg/kg~51.93 µg/kg) to 18.80 µg/kg (15.95 µg/kg~23.49 µg/kg), probably due to the increased biodegradation and photodegradation as winter shifted to spring. However, a comparison of results for the vegetable planting ponds and control ponds showed similar effects (56% and 59%, respectively; p = 0.460) in antibiotic reduction. The results suggested that the shading of the stems and leaves does not play a vital role in the reduction in total antibiotics during the complete cultivation cycle. The initial levels of total antibiotics in sediments were 46.82 µg/kg for the control ponds, 44.16 µg/kg for the broccoli ponds, and 43.35 µg/kg for the potherb mustard ponds. However, in the prior period, the ponds with broccoli and potherb mustard growth generated slightly higher antibiotic concentrations (36.19 µg/kg and 31.79 µg/kg) than those of the control pond (26.95 µg/kg), which decreased by 18%, 27%, and 42%, respectively. We doubt that photodegradation was impeded by vegetables above the sediment [27–29]. In the later period, 40%, 48%, and 30% of antibiotics were degraded in the potherb mustard pond, broccoli pond, and control pond, respectively. As such, antibiotic concentrations in the vegetable planting ponds declined faster than those in the control ponds, which may be due to the change in root environment and better degradation of antibiotics.

![Figure 2. Antibiotic concentrations in the sediments of labeled aquaculture ponds as shown in Figure 1.](image-url)
The studied ponds in Hangzhou Bay had a medium level of antibiotics in the sediment compared with other aquaculture ponds in China. For example, the total antibiotic amount in the aquaculture sediment in the Pearl River estuary (Zhuhai and Daya Bay) ranged from $7.99 \, \text{g/kg}$ to $22.73 \, \mu\text{g/kg}$ [30]. In addition, the values detected in the tilapia culture area in Guangxi Province varied from $3.22 \, \mu\text{g/kg}$ to $83.2 \, \mu\text{g/kg}$ and averaged at $28.3 \pm 27.5 \, \mu\text{g/kg}$, which is higher than that in the estuary and coastal sediment (16 ± 4.1 µg/kg) [31]. Moreover, the reported antibiotic levels in the sediment of the tested aquaculture area were $42.683 \, \mu\text{g/kg}$, $35 \, \mu\text{g/kg}$, and $106.94 \, \mu\text{g/kg}$ in Guangxi Province, Guangdong Province, and Tianjin, respectively [32–34].

### 3.2. Speciation Evolution of Antibiotics in the Shrimp–Vegetable Rotational Farming Period

Figure 3a shows the target antibiotics speciation in the ponds with TMP, OTC, and NOF (norfloxacin) as the most abundantly detected and accounting for 90% of the total antibiotic amounts. The respective levels of the detected antibiotics changed with the testing period as shown in Figure 3b for TMP, OTC, and NOF. For instance, TMP exhibited the greatest concentration in each pond (29.33 µg/kg in L-3 ponds, 27.06 µg/kg in K-2 ponds, and 27.04 µg/kg in X-1 ponds), but decreased quickly in March by 40–50%. TMP and OTC became the most dominant antibiotics and also declined in April. Although sulfadiazine, sulfamethazine, chlortetracycline, and roxithromycin were frequently detected (exceeding 80% of the sediment samples), their concentrations were relatively low. In contrast, only half of the sediment samples were found to have sulfadiazine, sulfamethoxazole, and erythromycin with no single detection of tetracycline.

![Figure 3. (a) Compositions of the analyzed antibiotics in each pond during the improvement period; (b) TMP concentrations in the studied ponds during the improvement; (c) OTC concentrations in the studied ponds during the improvement; (d) NOF concentrations in the studied ponds during the improvement.](image-url)
OTC is a broad-spectrum antibiotic in the tetracycline group. Figure 3c shows the average OTC concentrations of potherb mustard (X-ponds), broccoli (L-ponds), and the control ponds (K-ponds) were 16.82 µg/kg, 16.98 µg/kg, and 17.35 µg/kg, respectively. Unlike TMP, OTC had minor changes before March and significantly decreased in April. The enhanced degradation rate of OTC is likely attributed to direct photodegradation [16]. In addition, the plant roots may have changed the properties and structure of the sediment and promoted biodegradation. Figure 3d shows that the NOF level fluctuated without apparent seasonal dependence. NOF is an amphoteric ion with carboxyl, carbonyl, and amino groups that affect its adsorption and partitioning in sediment. The fluctuation probably resulted from altered microenvironments after plant growth that may have changed the sediment pH and NOF sorption behavior [35].

3.3. Antibiotic Characteristics after a New Round of Shrimp Farming

Figure 4 shows that antibiotics levels dramatically increased after a new round of Litopenaeus vannamei shrimp cultivation, to 52.70 µg/kg, 50.75 µg/kg, and 55.14 µg/kg, respectively, in the potherb mustard (X-ponds), broccoli (L-ponds), and control ponds’ (K-ponds) sediment. This major increase was mainly due to the addition of feed during the new round of cultivation. Table 4 compares the levels of MP, NOF, and OTC in pond sediment before and after the new round of shrimp cultivation, which indicates a multiple increase in those major antibiotic species in the sediment.

![Figure 4. Distribution of antibiotic species and their abundance in pond sediment.](image)

Table 4. TMP, NOF, and OTC content in pond sediment before and after a new production and culture circle.

| Antibiotic            | Potherb Mustard (X-Ponds) | Broccoli (L-Ponds) | Control Ponds (K-Ponds) |
|-----------------------|---------------------------|--------------------|-------------------------|
|                       | TMP | NOF | OTC | TMP | NOF | OTC | TMP | NOF | OTC |
| Before shrimp cultivation | 12.88 | 1.61 | 3.78 | 12.86 | 1.86 | 3.18 | 11.61 | 2.14 | 4.33 |
| After shrimp cultivation (µg/kg) | 15.18 | 7.91 | 27.19 | 13.49 | 8.57 | 26.87 | 17.84 | 8.37 | 26.98 |
| Increments (µg/kg)     | 2.3 | 6.3 | 23.41 | 0.63 | 6.71 | 23.69 | 6.23 | 6.23 | 22.65 |
| Increments (%)         | 18 | 391 | 619 | 5 | 361 | 745 | 54 | 291 | 523 |
4. Conclusions

Through a complete cultivation-cycle field investigation, this study found that the total antibiotic content decreased in the long term by 56% and 59% in the vegetable-growing shrimp ponds vs. the control ponds, respectively. The total amount of antibiotics in the traditional control decreased faster than that in the new aquaculture method in the prior period, which may have caused an interference in the photodegradation due to the vegetable growth. There was no significant difference in the total amount of antibiotics in each pond, and the stem and leaf shelter of vegetable planting did not greatly affect the reduction in the total antibiotic amount. The highest antibiotic contents in each pond were from TMP, OTC, and NOF, which accounted for more than 90% of the total. There were significant differences in the degradation process of the main antibiotics. The traditional control pond was more conducive to the photodegradation of TMP due to the higher TMP residuals in the vegetable growing ponds. However, the OTC content in the traditional control pond was higher than that in the vegetable-growing ponds. The vegetable-modified pond better facilitated the degradation of OTC, as the growing vegetables may have changed sediment properties and microbial communities that promoted the degradation of oxytetracycline. It was indicated that the system remediated the shrimp pond ecosystem as well as providing the possibility of increasing profits by planting vegetables in the winter idle period of shrimp ponds. Due to the ecological and health risks of antibiotics in the ponds, additional experiments should be conducted to elucidate the mechanisms of such aquaculture approaches to reduce antibiotics and sustainably improve shrimp yield.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/su13158400/s1, Table S1. Total concentration of Sediment Antibiotics in the Shrimp–Vegetable Rotational Farming Period. (µg/kg), Table S2. Concentration of each Sediment Antibiotics in the Shrimp–Vegetable Rotational Farming Period. (µg/kg).

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