Migration Characteristics and Quantitative Model of Heavy Metals in the Soil-Rice System in Typical Rice-Producing Areas of Zhejiang Province: A Postprint

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Abstract

The migration and transformation patterns of heavy metals in soil-rice systems have garnered increasing attention in the scientific community. This study collected soil and rice samples from three major rice-producing regions in Zhejiang Province at different locations (Nanxun in northern Zhejiang, Shengzhou in central Zhejiang, and Wenling in southern Zhejiang), conducted analysis of factors influencing heavy metal migration and transformation based on rice varieties (hybrid rice and late japonica rice), identified the primary driving factors, established a quantitative evaluation model and validated it, thereby characterizing the migration and transformation patterns of heavy metals in typical rice-producing areas of Zhejiang Province. The results showed that there were differences in soil physicochemical properties among the three study areas, wherein the pH (mean value of 5.52), organic matter (mean value of 39.4 $g \cdot kg^{-1}$), and heavy metal speciation content of soils in the Shengzhou production area were all lower than those in the other two study areas. Significant differences in heavy metal content were also observed in soils and rice among the three production areas, with soil heavy metal (Cd, Cu, Zn) content in the Wenling production area being significantly higher than in the other two areas ($P<0.05$), while rice heavy metal content in the Shengzhou production area was significantly higher than in the Nanxun area ($P<0.05$). The enrichment coefficients of Cd and Zn were higher than those of Cu and Ni, and the heavy metal enrichment coefficients in the Shengzhou production area (0.018–0.521) were significantly higher than those in the other two areas (0.004–0.143 for Nanxun, 0.007–0.269 for Wenling). Both soil heavy metal speciation and soil physicochemical properties significantly affected the enrichment coefficients of different varieties, with soil physicochemical properties having a relatively greater effect. The log-linear migration model could predict the availability (enrichment coefficient) of heavy metals in the soil-rice system under actual field conditions, with prediction results for late japonica rice being better than those for hybrid rice. The predictive performance for heavy metal Ni (regression coefficient r reaching 0.61 and 0.70 for hybrid rice and late japonica rice, respectively, $P<0.01$) was better than that for other heavy metals, while the predictive performance for heavy metal Cd in hybrid rice (r value of 0.21, $P>0.05$) was relatively low, requiring additional environmental variables and further research to improve prediction accuracy.

Full Text

Preamble

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Characteristics and Quantitative Model of Heavy Metal Transfer in Soil-Rice Systems in Typical Rice Production Areas of Zhejiang Province*

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Abstract

Understanding the characteristics of heavy metal transfer in soil-rice systems can improve soil quality in production areas and guide the safe production of rice. We collected soil and rice samples from three typical rice production areas (Nanxun, Shengzhou, and Wenling) located in the northern, central, and southern parts of Zhejiang Province. The controlling factors of heavy metal transfer were studied based on a transfer model set up for hybrid rice and japonica rice. The objective of the study was to identify transfer traits of heavy metals in soil-rice systems in typical rice production areas in Zhejiang Province and to guide safe agricultural production. The results suggested that the physico-chemical properties were different in the three areas. pH (mean value of 5.52), organic matter (mean value of 39.4 $g \cdot kg^{-1}$), EC and heavy metal fractions contents in soil in Shengzhou area were lower than those in the other two production areas. Sand content of soil in Shengzhou area was higher than that in the other two areas. Heavy metals in soils and rice were significantly different from each other of rice production areas. Heavy metals (Cd, Cu and Zn) contents in soil in Wenling area were significantly higher than those in the other two areas. Then heavy metals contents in rice in Shengzhou area were significantly higher than those in Nanxun area ($P < 0.05$). No carbonate bound fraction of heavy metals was detected in the study. The corresponding contents of exchangeable, Fe-Mn oxide bound, organic bound, and residual fractions of heavy metals in Shengzhou were lower than those in the other two production areas due to the lowest total heavy metals contents in Shengzhou soil. The enrichment indexes (EI) of heavy metals were different in the three production areas. Generally, EIs of Cd and Zn were higher than those of Cu and Ni. Also EIs in Shengzhou area (range of 0.018–0.521) were significantly higher than those in the other two areas (range of 0.004–0.143 for Nanxun area and of 0.007–0.269 for Wenling area). Both soil physico-chemical properties and heavy metals fractions were important factors influencing heavy metal enrichment indexes. Compared with heavy metals fractions, soil physico-chemical properties contributed more to the movement of heavy metals in soil-rice systems. A log-linear model of heavy metals combined with the physico-chemical properties and heavy metal fractions well predicted the availability of heavy metals in soil-rice systems under practical production conditions. The accuracy of the model prediction for hybrid rice was better than that for japonica rice. The Ni (regression coefficient r was 0.61 and 0.70 at $P < 0.01$ for hybrid and japonica rice, respectively) model was better than that of other heavy metals. However, the accuracy of the model prediction of hybrid rice Cd content ($r = 0.21$ at $P > 0.05$) was poor. In that case, it was necessary to conduct further research in order to improve the accuracy of the model by either using more of the environmental variables or adjusting the variables.

Keywords: Soil-rice system; Heavy metal; Transfer model; Controlling factor; Enrichment index; Risk evaluation

Introduction

Soil serves as the direct medium for crop growth, and consequently, toxic and harmful substances in soil can enter agricultural products through crop uptake. Among various pollutants, soil heavy metals have attracted widespread attention due to their persistence and resistance to biodegradation. Heavy metals in soil are absorbed by crop roots and accumulate in plant tissues. When their concentrations exceed the maximum permissible levels in agricultural products, food safety and quality cannot be guaranteed. Therefore, the extent of soil contamination directly affects the quality of agricultural products.

Reports indicate that approximately 12 million tons of grain are contaminated by heavy metals annually in China and cannot be consumed. Soils in suburban areas of many cities have suffered varying degrees of heavy metal pollution. Once heavy metals entering soil through various pathways exceed the soil environmental capacity, they not only inhibit crop growth and development, leading to yield reduction, but also cause excessive heavy metal accumulation in agricultural products such as grains, vegetables, and fruits. More critically, toxic heavy metals can be transferred to humans through the food chain, posing serious threats to human life and health.

To date, numerous studies have investigated heavy metal pollution in soil-rice systems. Research results demonstrate that heavy metal transfer and transformation in these systems are influenced by multiple factors, including soil physicochemical properties, rice physiological characteristics, and pollutant concentrations. Ren Yanhong monitored heavy metal pollution in a soil-rice system near the Baoshan mining area and concluded that agricultural heavy metal pollution was extremely severe in the mining region, with rice heavy metal contents showing significant correlation with soil heavy metal concentrations. Different rice cultivars exhibit significant differences in heavy metal transfer and transformation. Wang Heng studied rice production areas in the Songhua River Basin of Northeast China affected by sewage irrigation and found that heavy metal pollution primarily originated from human activities, with linear models effectively predicting Pb, Cu, and Ni elements, while logarithmic models were more suitable for Cr. In actual field environments, soils are subject to diverse constraints, with complex spatial variations in soil and plant characteristics, and both heavy metal pollution levels and soil physicochemical properties exhibit significant spatial heterogeneity. Therefore, it is necessary to investigate heavy metal transfer patterns in soil-rice systems under actual production conditions.

This study focused on typical production areas in Zhejiang Province, collecting sufficient soil and rice samples. Using rice genotype as the modeling basis, we investigated the effects of soil physicochemical properties and heavy metal fractions on heavy metal transfer and transformation in soil-rice systems, evaluated transfer models for various heavy metals, and provided a foundation for guiding safe rice production in actual production areas.

1. Materials and Methods

1.1 Study Area and Sampling

Three typical rice production areas were selected: Nanxun (120°04 -120°29 E, 30°38 -30°56 N) in northern Zhejiang, Shengzhou (120°28 -121°07 E, 29°20 -29°50 N) in central Zhejiang, and Wenling (121°10 -121°44 E, 28°13 -28°32 N) in southern Zhejiang [Figure 1: see original paper]. All three study areas have a subtropical monsoon climate with distinct seasons, mild and humid conditions, an average annual temperature of approximately 16°C, and average annual precipitation of about 1,300 mm. Nanxun is dominated by conventional japonica rice, Shengzhou by hybrid indica rice, and Wenling has mixed cultivation of both conventional japonica and hybrid indica rice. The soils in Nanxun are primarily paddy soil, fluvo-aquic soil, and red soil; Shengzhou features red soil, yellow soil, lithosol, fluvo-aquic soil, and paddy soil; and Wenling consists mainly of paddy soil, red soil, skeletal soil, and coastal saline soil.

During the rice harvest season, sampling sites were selected using GPS navigation, considering different soil types and sampling distribution uniformity. Within a 10-meter radius circular area at each sampling site, five subsamples were collected and combined as the representative sample for that site. A total of 100, 94, and 96 paired samples of paddy soil and corresponding rice grains were collected from Nanxun, Shengzhou, and Wenling, respectively. The specific sampling locations are shown in Figure 1.

1.2 Sample Analysis

Soil samples were air-dried at room temperature and ground to pass through a 2-mm nylon sieve for physicochemical property analysis. A portion of these sieved samples was further ground in an agate mortar to pass through a 100-mesh sieve and stored for subsequent analysis.

Soil physicochemical properties were determined using conventional analytical methods. Total heavy metal concentrations in soil were measured using HF-HNO_3-$HClO_4$ tri-acid digestion. Heavy metal contents (Cu, Ni, Pb, and Zn) were determined by flame atomic absorption spectrophotometry (FAAS, Perkin Elmer AA800, USA), while Cd content was measured by graphite furnace atomic absorption spectrophotometry (GFAAS, Perkin Elmer AA800, USA).

Heavy metal fractionation in soil was performed using the widely adopted Tessier sequential extraction procedure, which extracts exchangeable, carbonate-bound, Fe-Mn oxide-bound, organic-bound, and residual fractions. Soil Cd fractions, as well as exchangeable and organic-bound Ni, were determined by GFAAS, while other metal fractions were analyzed by FAAS.

Rice plant samples were oven-dried at 105°C for 1 hour to deactivate enzymes, then dried at 70°C to constant weight. The samples were subsequently husked, polished, ground, and stored in sealed plastic bags. Rice heavy metals were digested using HNO_3-H_2O_2. Zn content was determined by FAAS, while Cd, Cu, Ni, and Pb contents were measured by GFAAS.

1.3 Data Analysis and Transfer Model

Since Pb concentrations in rice were below the instrument detection limit, statistical analysis and modeling for Pb in the soil-rice system were not conducted in this study. Descriptive statistical analysis, normality tests, variance analysis for significant differences, correlation significance tests, and model analyses were all performed using SPSS 18.0 software.

Shengzhou was selected as the study area for hybrid rice, while Nanxun was chosen for conventional japonica rice. The heavy metal enrichment index (EI), defined as the ratio of heavy metal concentration in rice to that in soil, was used to represent transfer capacity. Soil heavy metal fractions and physicochemical properties were considered as influencing factors.

Previous studies have shown that correlation models between soil and crop heavy metals primarily include linear, exponential, and logarithmic models. When soil heavy metal concentrations are relatively low, linear relationships are typically observed. Exponential and logarithmic models are mathematically interconvertible, allowing soil-crop system heavy metal models to be categorized into linear and log-linear models.

Accordingly, both linear and log-linear fittings were performed for each heavy metal for the two rice cultivars, and the model with the best fit was selected. The optimal transfer model for the soil-rice system heavy metals in this study is expressed as:

$$\ln EI = a\ln I + b\ln II + c\ln III + d\ln IV + epH + fOM + gEC + h\text{Sand} + i\text{Silt} + j\text{Clay} + k$$

where *EI* is the heavy metal enrichment index; I, II, III, and IV represent exchangeable, Fe-Mn oxide-bound, organic-bound, and residual fractions of heavy metals, respectively; *a, b, c, ⋯, k* are model regression coefficients; *OM* is soil organic matter content; *EC* is electrical conductivity; and Sand, Silt, and Clay represent the contents of sand, silt, and clay particles, respectively.

2. Results and Analysis

2.1 Physicochemical Properties of Paddy Soils

Soil physicochemical properties varied among the three production areas. Shengzhou soils had lower mean pH (5.52) and organic matter content (39.4 $\text{g} \cdot \text{kg}^{-1}$) compared to Nanxun and Wenling . Additionally, this area exhibited lower electrical conductivity, silt, and clay contents, while sand content was significantly higher ($P < 0.05$) than in the other two areas. These differences in soil properties contributed to variations in heavy metal enrichment coefficients among the production areas.

2.4 Comparison of Heavy Metal Enrichment Coefficients in Soil-Rice Systems

Heavy metal enrichment coefficients in the soil-rice system showed similar distribution patterns in Shengzhou and Wenling [Figure 2: see original paper]. In both areas, Cd had the highest enrichment coefficients (0.521 and 0.269, respectively), followed by Zn (0.245 and 0.160), while Cu and Ni had relatively lower coefficients. In Nanxun, Zn exhibited the highest enrichment coefficient (0.143), followed by Cu, Cd, and Ni.

Significant differences in enrichment coefficients were observed among the three production areas. Variance analysis revealed that enrichment coefficients in Shengzhou were significantly higher ($P < 0.05$) than those in Nanxun and Wenling. Nanxun and Wenling also showed significant differences ($P < 0.05$) in Cd and Ni enrichment coefficients, while no significant differences were found for Cu and Zn. Consequently, the heavy metal transfer capacity in soil-rice systems across the three areas followed the order: Shengzhou > Wenling > Nanxun.

2.5 Prediction Model Analysis

Using soil heavy metal fractions, physicochemical properties, and their combinations, enrichment coefficients for different rice cultivars were predicted with the best-fit models . The goodness-of-fit (R) varied with rice cultivar and heavy metal. For Cd, the combined model of fractions and soil properties achieved R values of 0.65 and 0.56 for hybrid rice, and 0.50 and 0.76 for japonica rice, all reaching highly significant levels. Notably, soil properties alone showed better fit than metal fractions for japonica rice. Both fractions and soil properties achieved highly significant fits for hybrid rice Cu, while soil properties alone showed highly significant fit for japonica rice Cu, with fractions showing only significant fit. For Ni, fractions showed non-significant fit for hybrid rice ($R = 0.20$), whereas soil properties achieved highly significant fit; both fractions and properties showed highly significant fits for japonica rice. All Zn transfer models achieved highly significant fit levels, indicating that Zn transfer in both cultivars was influenced by Zn fraction contents and soil physicochemical properties.

Regression coefficients for each factor in the combined model of heavy metal fractions and soil physicochemical properties are presented in . The transfer models were validated using both rice cultivars from Wenling area [Figure 3: see original paper]. Except for Cd in hybrid rice, correlation coefficients between measured and predicted values for all other heavy metals reached highly significant levels. For Cd, hybrid rice showed poor predictive capability ($r = 0.21$, $P > 0.05$), while japonica rice achieved $r = 0.52$ ($P < 0.01$). When enrichment coefficients were low, model predictions overestimated measured values; as enrichment coefficients increased, predictions underestimated measured values, with distinct threshold points at 0.2 for hybrid rice and 0.1 for japonica rice. Cu predictions for both cultivars were highly significant, though hybrid rice predictions tended to underestimate measured values with increasing deviation at higher enrichment coefficients. Ni showed the best prediction results, with model predictive capabilities of 0.61 and 0.70 for hybrid and japonica rice, respectively ($P < 0.01$). Zn predictive capability was comparable to Cu, though both cultivar models overestimated measured values at low enrichment coefficients.

3. Discussion and Conclusion

Soil physicochemical properties play crucial roles in heavy metal transfer and transformation. In this study, the low pH, low organic matter content, and low electrical conductivity in Shengzhou promoted heavy metal transfer and uptake in the soil-rice system. Conversely, higher soil pH, electrical conductivity, and silt-clay textures in Nanxun and Wenling were less conducive to heavy metal enrichment. Therefore, despite having the lowest total heavy metal concentrations among the three areas, Shengzhou rice contained the highest heavy metal levels, indicating higher bioavailability and potential heavy metal pollution risk in the Shengzhou soil-rice system. This also demonstrates that total heavy metal content cannot adequately characterize heavy metal bioavailability.

Enrichment coefficients for Cd and Zn were higher than those for Cu and Ni, indicating relatively higher transfer capacity and bioavailability of Cd and Zn in the soil-rice system. Non-residual fractions (exchangeable, Fe-Mn oxide-bound, and organic-bound) of soil Cd and Zn exhibited certain bioavailability to rice.

These fractions constituted high proportions of total soil content in the study areas. For example, non-residual Cd accounted for 91.6%, 90.2%, and 92.8% of total Cd in Nanxun, Shengzhou, and Wenling, respectively. In contrast, exchangeable Ni, the bioavailable form for rice, was extremely low, representing only 1.8%, 2.5%, and 4.7% of total soil Ni. These findings are consistent with previous studies. Zhu et al. reported that rice Cd content did not significantly correlate with total soil Cd but showed good correlation with Cd extracted by various extractants. Wang Heng observed that enrichment coefficients for Zn and Cd were significantly higher than those for Cu, Ni, and other elements.

This study successfully fitted relationships between enrichment coefficients and soil heavy metal fractions and physicochemical properties using combined linear and log-linear models. Wang et al. used linear models to predict Pb, Cu, and Ni contents in rice and log-linear models for Cd and Cr. Overall, models based on soil physicochemical properties outperformed those based on heavy metal fractions. For Ni, soil properties achieved highly significant fits for both cultivars, while metal fractions showed non-significant fit for hybrid rice and lower coefficients for japonica rice than soil properties. This indicates that soil physicochemical properties drive heavy metal transfer more strongly than heavy metal fraction contents. Among the three model types, the combined fraction and property models achieved the highest goodness-of-fit, with both cultivar models reaching highly significant levels.

Although the fitted models achieved significant levels, their determinant coefficients R (maximum = 0.83) were lower than those from previous pot and plot experiments. This is because actual rice production areas represent complex ecosystems with spatial heterogeneity in soil heavy metal content, physicochemical properties, and management practices. This study focused on effects of heavy metal fractions and soil properties (pH, organic matter, electrical conductivity, particle composition), while other factors such as soil redox potential and phosphorus content also influence heavy metal bioavailability. Incorporating these factors could improve model performance. Additionally, transfer models varied with rice cultivar and heavy metal type. These models could adequately predict heavy metal bioavailability (enrichment coefficients) in actual production environments, though predictive capability differed by cultivar and metal. Japonica rice predictions were better than hybrid rice, and Ni predictions outperformed other metals. However, the models tended to overestimate low enrichment coefficients with relatively large local errors, particularly for Cd. Therefore, the cultivar- and metal-specific transfer models developed in this study require further improvement and calibration to enhance predictive accuracy and practical applicability.

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