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Interaction of Soil Nutrients and Arsenic (As) in Paddy Soil in a Long-Term Fertility Experiment

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Abstract: In this study, we examined the interaction between arsenic (As) and nutrients in paddy soil which received pig manure and chemical fertilizers for 36 years (since 1984). The treatments consisted of: CK (without fertilization); NPK (chemical nitrogen, phosphorus and potassium fertilization); NPK30%M (70% NPK plus 30% manure); NPK50%M (50% NPK plus 50% manure); and NPK70%M (30% NPK plus 70% manure). The combined application of pig manure and chemical fertilizer improved grain yield, soil pH and nutrient levels compared to chemical fertilizer application treatment. In comparison to CK, grain yield increased by 55.9%, 75.0%, 74.9% and 71.9%, respectively under the NPK, NPK30%M, NPK50%M and NPK70%M treatments. Soil As concentration increased by increasing the amount of manure input, and the highest concentration of As was 0.64 mg kg^{-1} found in the NPK70%M treatment. Increasing the rate of manure application decreased the As bioaccumulation coefficient (BAC) for rice grain. SOC, total N and P showed a positive correlation with the soil-available As concentration and negative correlation with BAC. Furthermore, the partial least square model (PLS) showed that the soil pH and SOC were the most influencing factors on BAC among the different properties of soil, which explained the 75.4% and 17.6% of total variations, respectively. This study concluded that the addition of pig manure together with chemical fertilizers can increase crop production by supplying essential nutrients, but the concentration of As in manure should be monitored to reduce soil and food contamination.

Keywords: arsenic; bioaccumulation of As; long-term fertilization; paddy soil; rice

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1. Introduction

Arsenic (As) contamination in environmental matrices is recognized as one of the serious threats to food security and human safety throughout the world [1,2]. According to an estimation, about 200 million people have suffered from As contamination either by consuming As-polluted drinking water and/or food such as rice (*Oryza sativa L.)* in which As has accumulated in the rice grains through As-contaminated irrigation water [3,4]. The main sources of As contamination in soil are rock weathering (natural source) and anthropogenic activities, such as pesticide application [4,5]. The mechanism of As uptake in rice grains is the conversion of arsenate As(IV) into more toxic arsenite As(III) under flooded conditions in paddy soil and its subsequent translocation and accumulation in rice grain [6,7]. According to the guidelines of the World Health Organization (WHO), the maximum safe level for As is 10 μg L^{-1} in water used for drinking purposes and 100 μg L^{-1} in water used for irrigation purposes [8].

As a staple food, rice is consumed by roughly 3 billion people worldwide, primarily in Asian countries, where it meets 70% of their energy needs. According to the Food and Agriculture Organization (FAO) guidelines, 0.2 mg kg^{-1} dry weight (DW) is a safe level of As in rice [9]. Rice is grown in around 115 nations around the world, and about half of the rice-growing countries in Asia have two to three harvests. [10]. It is only grown once a year in temperate regions, and the growing seasons vary by country. Rice crops are grown under flooded conditions, and in these submerged conditions, reduction reaction occurs which converts less toxic As(V) to the As(III) species, which are more toxic and phytoavailable [11,12]. Generally, the uptake of As by plant roots is higher in its inorganic form compared to its organic compounds [13,14]. Total As in noncontaminated soil ranged from 0.1–10 mg kg^{-1} [15]. According to the recommendations of the European Union (EU), the safe limit of total As in cultivated land is below 20 mg kg^{-1} [16].

The biogeochemical process in paddy soil favors the uptake of As in rice crops and the subsequent As buildup in rice grains [1]. Several parameters, including soil pH, redox potential (Eh), soil carbon, phosphate, sulphate, metal oxides and the microbial community structure influence As uptake in rice crops in paddy soil. [17].

Recently, a lot of interest has developed in investigating the impact of different organic and inorganic inputs on the uptake of As by rice crops in paddy fields [18,19]. Manure application to farm lands has become a common practice across the world to increase soil organic matter and other nutrients to achieve a higher agricultural output. Recently, the development of the pig industry has increased the production of pig manure. The addition of pig manure to soil as fertilizer improves soil structure and replenishes soil nutrients as well as assisting in the recycling of agricultural residues. On the other hand, pig manure can include hazardous components such as heavy metals that could be taken up by plants [13,20]. However, the application of manure can also influence the soil nutrient content and pH, which can have a direct or indirect influence on the uptake and bioaccumulation of As in rice crops [21]. Therefore, manure application may pose a risk of As pollution in soil and crops. Thus, it is essential to investigate the influence of pig manure treatments on As bioaccumulation in rice under field conditions. Moreover, previous studies on As uptake were mainly carried out for a short duration and in limited environmental conditions which produced contradictory results [22]. A limited number of studies have been undertaken to understand the behavior of soil nutrients and As uptake by crops under long-term fertilization. Therefore, in this study, we aimed to investigate the long-term impact of the application of pig manure and chemical fertilizers on crop productivity and As contamination. It is hypothesized that the addition of a high rate of pig manure application can increase the As contamination of soil and grain and that the change in soil nutrient content under long-term pig manure application can influence As accumulation in rice grains. The key objectives of the present research were to: (1) investigate the impact of long-term varying rates of manure application and inorganic fertilization on crop yield and bioaccumulation of As in rice crops and (2) investigate the interaction of nutrients and As in a rice-based cropping system.

2. Materials and Methods

2.1. Experimental Site

A long-term field trial was established in 1984 at a research field of the Jiangxi Academy of Agricultural Sciences (28°57′ N, 115°94′ E) in Nanchang County, China. The climate type at the farm was subtropical with mean annual precipitation of 1600-mm and annual temperature of 17.5 °C. Soil type was red paddy which is classified as ferralic cambisol [23] and cultivated with rice. Initial soil properties (at depth of 0–20 cm) comprised pH of 6.5, SOC of 14.9 mg kg^{-1}, total N (TN) of 1.4 g kg^{-1}, TP of 0.50 g kg^{-1}, available N (AN) of 172 mg kg^{-1}; AP of 21.0 mg kg^{-1} and AK of 35.1 mg kg^{-1}. Total mean concentration of As in irrigation water was 1.036 μgL^{-1} and in pig manure (dry weight), was 4.33 ± 0.10 mg kg^{-1}. Water samples for the determination of As content were taken from an irrigation channel near the field.

2.2. Experimental Setup and Crop Management

Randomized complete block design (RCBD) was followed to arrange the fertilizer treatments. Each treatment had three replications (plot size 1.9 × 16.0 m^2). To avoid the contamination from nearby plots, each plot was split from each other by cemented barriers. Fertilizer treatment levels for the rice crop within this area was based on local recommendation. The treatments included: (1) CK (without fertilization); (2) NPK (chemical N, P and K fertilization); (3) NPK30%M (70% NPK plus 30% pig manure); (4) NPK50%M (50% NPK plus 50% pig manure) and (5) NPK70%M (30% NPK plus 70% pig manure). Fertilizer N, P and K were supplied at the rate of 180 kg^{-1}, 28.5 kg ha^{-1} and 124.5 kg ha^{-1}, respectively. Pig manure input rate was 12000 kg ha^{-1} in NPK30%M, 20,022 kg ha^{-1} in NPK50%M and 2800 kg ha^{-1} in the NPK70%M treatment. Chemical fertilizers were applied as urea for N, calcium superphosphate for P and potassium chloride for K. Urea and potassium chloride were supplied in split form (50% as basal and 50% as topdressing), while complete dose of P fertilizer and pig manure were added in soil during the land preparation for cultivation. Nutrient contents in pig manure were as follow: N, 4.5 g kg^{-1}; P, 0.82 g kg^{-1}; and K, 4.98 g kg^{-1}. Locally dominant cultivar of rice was sown, and the cultivar was replaced every three years. In the year of sampling for current work (2017–2018), rice cultivar Chunguang-1 was grown, and rice hill spacing was kept at 20 cm^2. Irrigation level of water was kept at 5–10 cm above ground level before the crop matured. Other management techniques, such as pest control, were carried out in accordance with local farming practices. The crop was harvested after it reached full maturity, and the straw was removed.

2.3. Sampling and Chemical Analysis

In 2017, soil samples (at depth of 0–20 cm) were obtained by using a stainless-steel sampler at five randomly selected spots in airtight plastic bags in each treatment unit. The representative samples were transferred to the laboratory for further analysis. Apart from composite samples, soil was air-dried and sieved (through 0.2 mm) for chemical characterization. The pH of soil was measured in soil: water (1:25) suspension. SOM, TN and TP were measured according to oxidation method [24], Black [25] and Murphy and Riley [26], correspondingly. The contents of AN and AP in soil were measured using Lu et al. [27] and Olsen [28] techniques, respectively. To measure the total As concentration, 10 mL of (1:1, (v/v) was used to digest soil (0.2 g) for 2 h at 100 °C, and 0.101 M $CaCl_2$ solution was used to extract plant-available As using 1:10 soil: solution ratio (w/w) and shaking for 2 h [29]. As concentration in solution was measured using atomic fluorescence spectrometry (AFS-9130 Beijing Jitian Instrument Company, Beijing, China; AQSIQ 2008) from a Chinese company, and accuracy of measurement was verified using a reference standard soil (GBW 07429). Rice grain were manually separated from the straw, air-dried overnight and digested in the solution of HNO_3/H_2O_2 to obtain an As reading using ICP-MS [11]. Replicated samples of fresh pig manure were air-dried, ground and ashed using $MgO+Mg(NO_3)_2$ as drying agent in the muffle furnace at 550 °C for 6 h. Residue was dissolved using HCl. Replicated and spiked samples (GBW-08501 from National Research Centre for Standards, Beijing, China) were added for QA/QC. 5% HCl solution was used as a reagent solution, and As concentration in final solution and in irrigation water was analyzed using a hydrogen generation atomic fluorescence spectroscopy (AFS-2202, Haiguang Company, Beijing, China) [30]. The reference standards were included in every set of samples to ensure correct measurements.

2.4. Calculations

The bioaccumulation coefficient (BAC) was estimated to gain a full understanding of As bioavailability in soil [31]. It shows the amount of As transferred from soil to rice grain. The equation used to calculate the BAC (%) is given below:

$$BAC\ (\%) = \frac{C_{rice}}{C_x} \times 100$$

where, C_{rice} denotes the heavy metal concentration in rice, and C_x denotes the heavy metal in soil.

2.5. Data Analysis Tools

All the dataset was analyzed using one-way ANOVA test, followed by Tukey's LSD at $p \leq 0.05$ significance level using SPSS (for Windows, Version 27.0., IBM, Armonk, NY, USA) [32]. Regression analysis was carried out using Sigmaplot (Windows version 14.0). Correlation matrix was performed using JMP Pro 14 (SAS Institute, Cary, NC, USA). Partial least square (PLS) model was performed to predict the influence of different soil properties on bioaccumulation coefficient using JMP Pro 14 (SAS Institute, Cary, NC, USA).

3. Results

3.1. Long-Term Fertilization Impact on Soil Characteristics

The long-term application of pig manure and chemical fertilizers showed a significant impact on soil properties (Table 1). The different rates of pig manure in combination with inorganic fertilizers significantly increased soil pH in comparison to the control and inorganic treatments. Among all the treatments, the minimum pH of soil was 5.06 under the NPK treatment, and the maximum was 5.76 under the NPK70%M treatment. Similarly, addition of manure also significantly increased the SOC and other nutrient contents in comparison to inorganic treatments. In comparison to the control (CK), SOC increased by 28.3%, 51.4%, 68.3% and 76.2%; TN increased by 11.9%, 37.0%, 55.4%, and 66.7%; and TP increased by 150%, 225.7%, 381.2% and 405.2%, respectively, under the NPK, NPK30%M, NPK50%M and NPK70%M treatments, respectively. The addition of manure in combination with inorganic fertilizers also increased the AN and AP content in soil, and the highest concentrations of AN and AP were under the NPK70%M treatment. Among all the treatments, the total As concentration ranged from 6.07 mg kg^{-1} under the NPK30%M treatment to 8.36 mg kg^{-1} under the CK treatment. However, the available As concentration shot up when the manure input was increased, and the greatest concentration of plant-available As was 0.64 mg kg^{-1} under the NPK70%M treatment. The soil BD was changed by different fertilization treatments, and it was decreased by increasing the manure input. Soil BD ranged from 0.93 g cm^{-3} under the NPK70%M to 1.22 g cm^{-3} under the NPK treatment.

Table 1. Soil physiochemical characterization under long-term application of chemical fertilizers and pig manure.

Treatments *	pH	SOC (g kg^{-1})	TN (g kg^{-1})	TP (g kg^{-1})	AN (mg kg^{-1})	AP (mg kg^{-1})	tAs (mg kg^{-1})	aAs (mg kg^{-1})	Soil BD (g cm^{-3})
CK	5.48 ± 0.02 b	12.94 ± 0.08 c	1.35 ± 0.03 c	0.44 ± 0.01 e	89.79 ± 3.44 d	10.82 ± 0.40 d	8.36 ± 0.47 a	0.09 ± 0.002 d	1.19 ± 0.01 a
NPK	5.06 ± 0.02 c	16.60 ± 1.00 bc	1.51 ± 0.02 c	1.11 ± 0.04 d	134.23 ± 6.05 c	52.65 ± 4.12 c	7.01 ± 0.22 b	0.35 ± 0.01 c	1.22 ± 0.01 a
NPK30%M	5.35 ± 0.06 b	19.60 ± 1.41 ab	1.85 ± 0.20 b	1.45 ± 0.05 c	163.35 ± 12.90 b	91.44 ± 6.05 b	6.07 ± 0.45 c	0.56 ± 0.02 b	1.00 ± 0.05 b
NPK50%M	5.55 ± 0.23 ab	21.78 ± 3.29 a	2.10 ± 0.02 a	2.14 ± 0.01 b	173.39 ± 5.01 b	100.32 ± 9.95 b	6.44 ± 0.20 bc	0.55 ± 0.01 b	0.97 ± 0.01 bc
NPK70%M	5.76 ± 0.11 a	22.81 ± 4.23 a	2.25 ± 0.01 a	2.24 ± 0.01 a	198.33 ± 8.20 a	116.38 ± 10.1 a	6.54 ± 0.24 bc	0.64 ± 0.01 a	0.93 ± 0.01 c

Numerical values are means ± standard deviations. Different letters with mean values indicate the significant differences from each other at $p \leq 0.05$ in accordance with Tukey's LSD test. Number of replications = n = 3. * CK, no fertilization; NPK, inorganic N, P and K fertilization; NPK30%M, 70% of inorganic NPK and 30% of manure fertilization; NPK50%M, 50% of inorganic NPK and 50% of manure fertilization; and NPK70%M, 30% of inorganic NPK and 70% of manure application.

3.2. Annual Rice Crop Yield and Bioaccumulation of Arsenic in Grain

Fertilization significantly influenced annual crop yield and concentration of As in rice grains (Figure 1), while grain yield and grain As concentration did not significantly differ across the NPK30%M, NPK50% and NPK70%M treatments. Grain yield increased by 55.9%, 75.0%, 74.9% and 71.9%, and grain As concentration increased by 19.4%, 15.0%, 14.9% and 10.2%, respectively, under the NPK, NPK30%M, NPK50% and NPK70%M treatments compared to the control treatment. SOM, TN and total P content were positively correlated with annual crop yield, but soil bulk density and total As concentration were negatively correlated with crop yield (Figure 2). Increasing the manure input significantly decreased BAC compared to NPK. The highest BAC value was under the CK treatment, followed by NPK, NPK30%M, NPK50%M and NPK70%M, respectively (Figure 3).

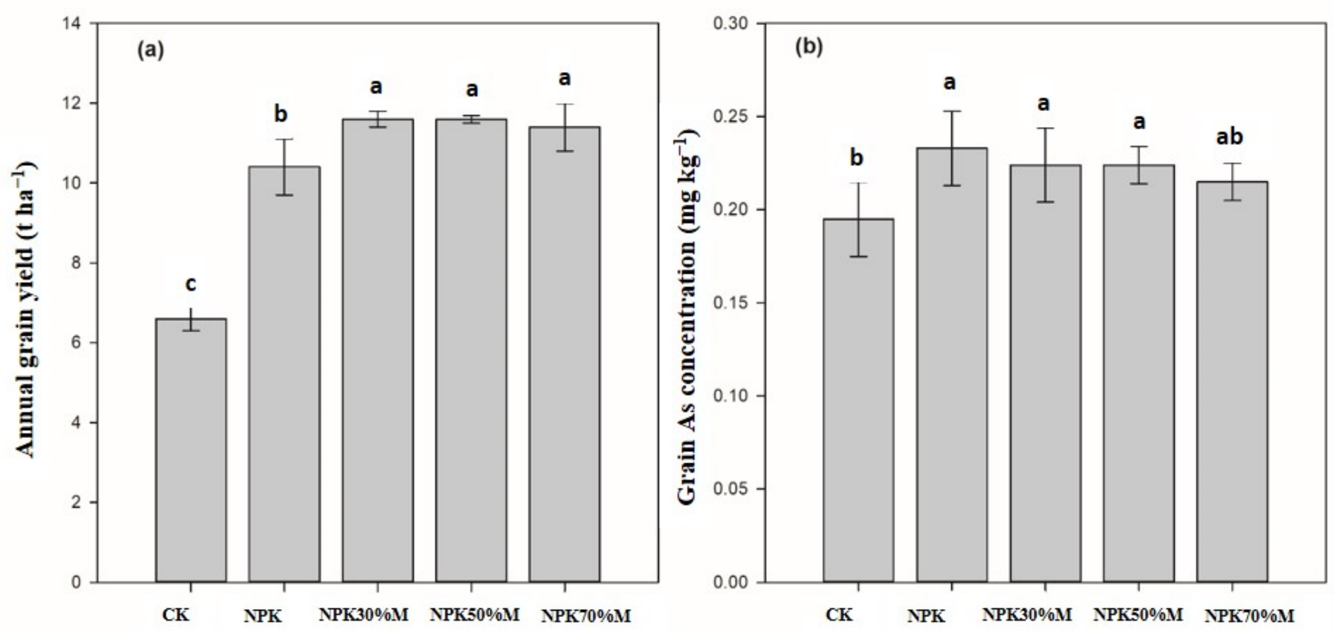

Figure 1. Annual rice grain yield (t ha^{-1}) (**a**) and arsenic (As) concentration (mg kg^{-1}) (**b**) in rice grains under application of chemical fertilizers and pig manure. Error bars denote ± standard deviations; over the bars, different letters indicate significant differences at $p \leq 0.05$ in accordance with Tukey's LSD test. Number of replications = n = 3.

3.3. Correlations between Bioaccumulation Coefficient of Arsenic and Soil Properties

In the correlation matrix, soil pH exhibited significantly ($p \leq 0.05$) positive relationships with SOC (r = 0.058), TN (r = 0.62) and TP (r = 0.51) (Figure 4). SOC also showed significant positive relationships with TN (r = 0.86), TP (r = 0.87), AN (r = 0.91), AP (r = 0.92) and available As (r = 0.85), and SOC showed negative correlations with soil total As (r = 0.73), BAC (r = 0.75) and BD (r = 0.74). Similarly, BAC was negatively correlated with TN (r = 0.75), TP (r = 0.84), AN (r = 0.88) and AP (r = 0.89), while soil BD showed significant negative correlations with soil pH, SOC, TN, TP, AN and AP, and BD showed positive relationships with BAC and the total As concentration in soil.

The partial least square regression (PLS) showed that soil pH was the most influencing factor on BAC, which accounted for 75.4% of the relative influence (Figure 5). The relative influence of SOC on BAC was 17.6%, while, TN, TP, AN, AP, AK and BD accounted for >5%.

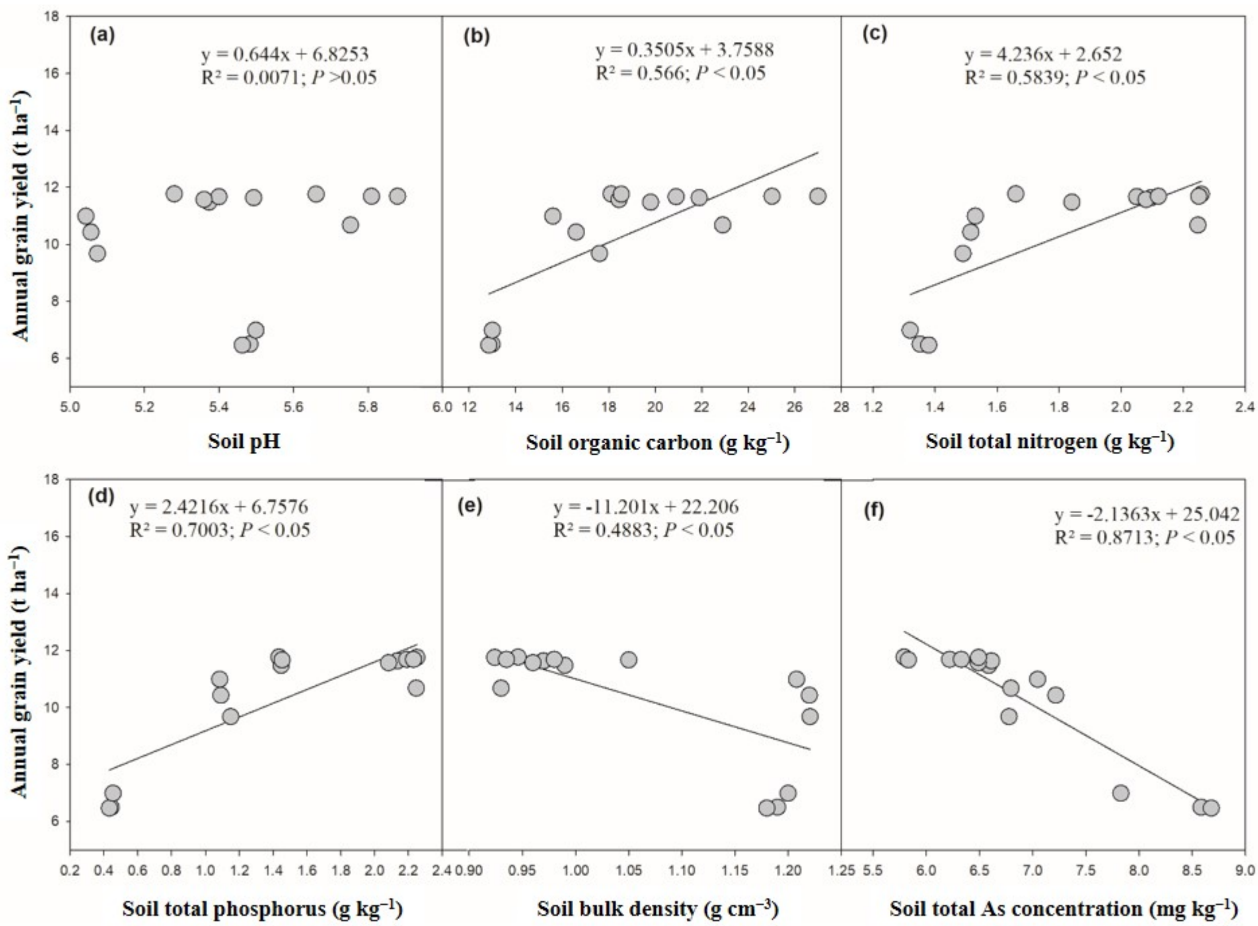

Figure 2. Linear regression analysis between grain yield and soil properties: (**a**) pH, (**b**) organic carbon (SOC), (**c**) total nitrogen (TN), (**d**) total phosphorus (TP), (**e**) bulk density and (**f**) soil total arsenic concentration (tAs).

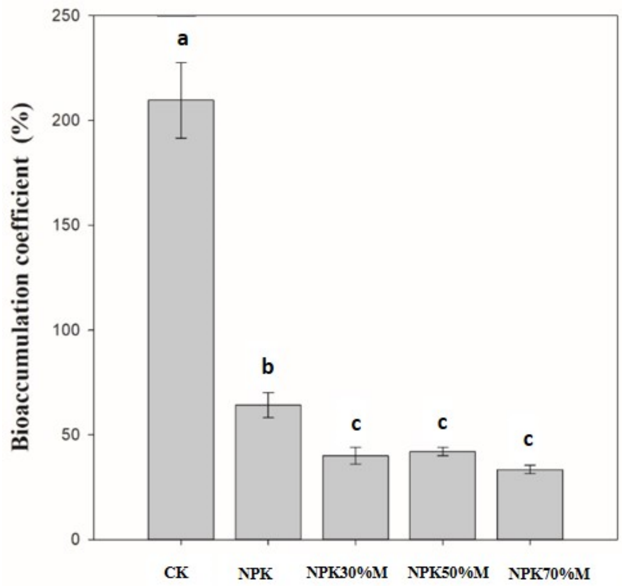

Figure 3. Arsenic bioaccumulation coefficient (BAC). Error bars denote ± standard deviations; over the bars, different letters indicate significant differences at $p \leq 0.05$ in accordance with Tukey's LSD test. Number of replications = n = 3.

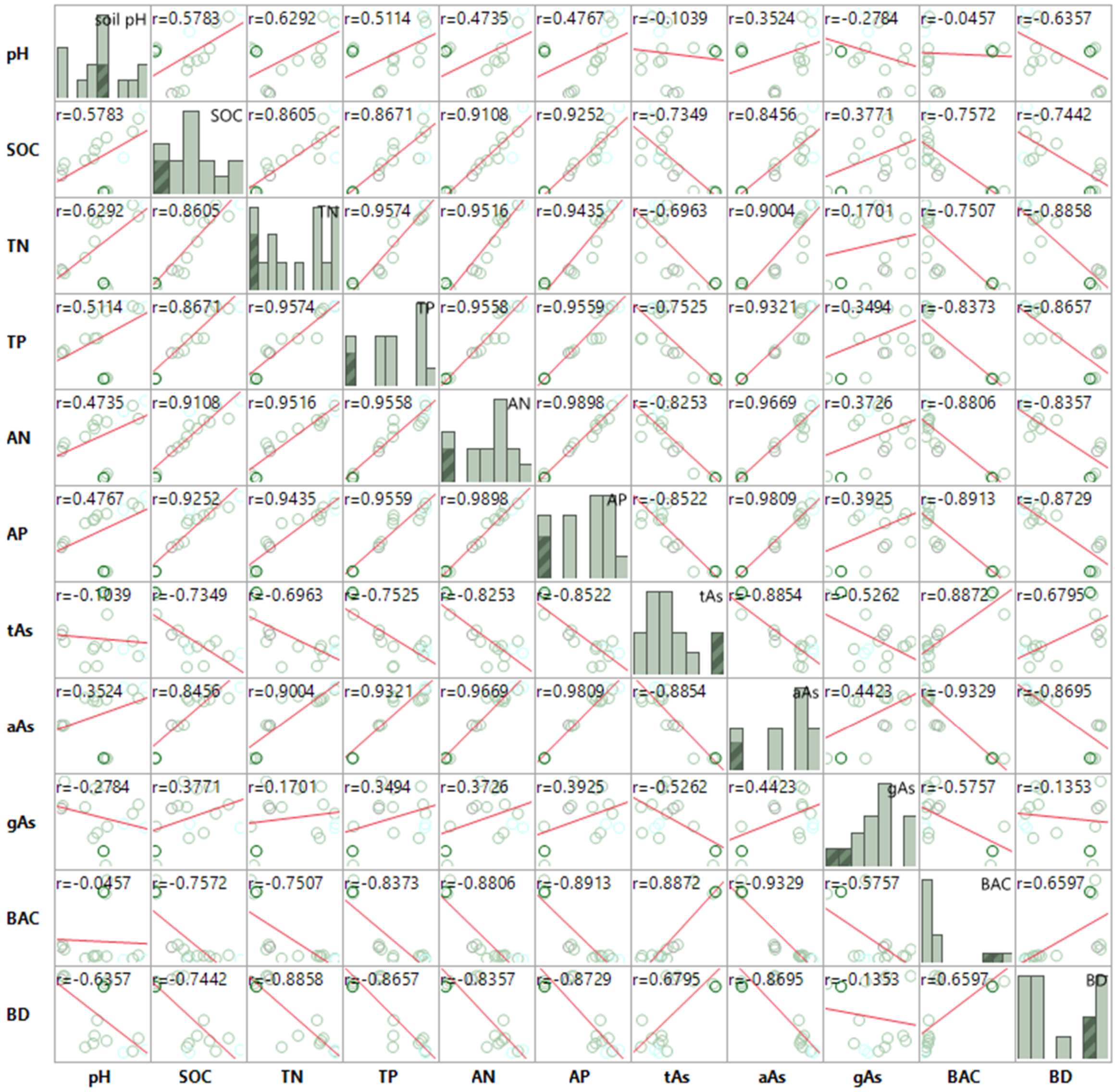

Figure 4. Correlation matrix among different soil properties and bioaccumulation coefficient (BAC).

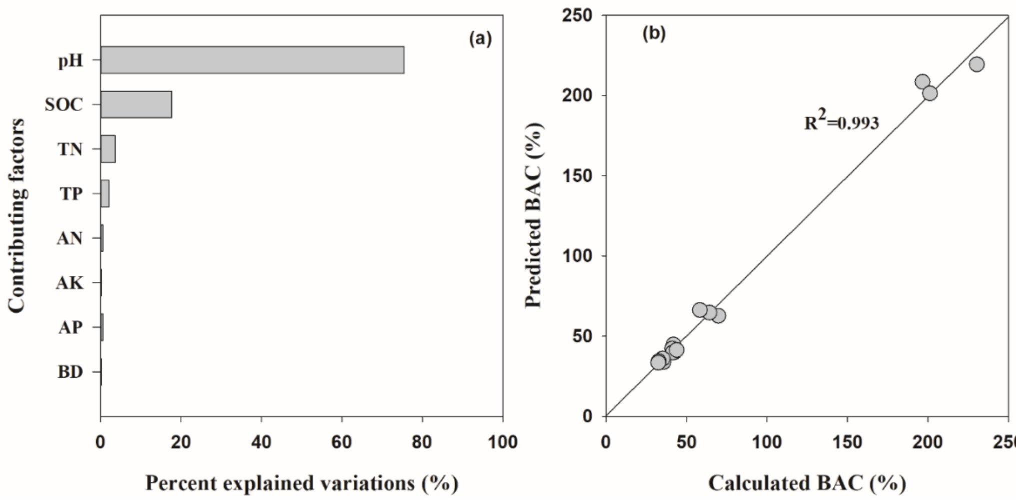

Figure 5. Partial least square regression showing percent explained variations for bioaccumulation coefficient (BAC) (**a**) and linear relationship between calculated BAC and predicted BAC (**b**).

4. Discussion

Organic amendments contain essential plant nutrients; therefore, the prolonged application of manures on farm land is considered as a promising and cost-effective reserve of plant nutrients to obtain a high crop yield [33–36]. Pig manure addition to agricultural land is encouraged by the Government of China, and its application to cropland has become a conventional practice due to the rapid growth of the pig industry in the country [37–40]. However, pig manure is also becoming a potential source of toxic and nonessential elements in agricultural soils [20,30]. Therefore, continuous pig manure addition may contaminate food crops through the bioaccumulation of toxic elements such as As in rice grains [4,11]. In the current research, continuous pig manure combined with inorganic fertilizer application notably affected the soil properties (Table 1) and crop yield (Figure 1). Soil pH, SOC, TN and TP contents were enhanced by increasing the amount of pig manure input (Table 1). These findings are in line with prior research, which found that applying a continuous combination of manure coupled with inorganic amendments enhanced the soil nutrient content and pH [41,42]. Manure is alkaline in nature due to the presence of a high concentration of alkaline elements such as Ca, K and Mg contents; therefore, it increased the soil pH in the paddy soil [43,44]. Manure alkalinity might also be due to ammonia nitrogen production from organic N and anion decarboxylation, which increases the soil pH [45]. On the other hand, mineral N input reduces the base cation saturation ratio in soil and decreases the pH; therefore, the soil pH was lowest under the NPK treatment in this study. Similarly, crop yield was significantly higher under the combined application of inorganic fertilizers and pig manure compared to the NPK treatment (Figure 1). This was due to the high presence of carbon and nutrient content under the combined manure and chemical fertilizer application in comparison to chemical fertilization. These findings are in line with those of earlier long-term fertility experiments [46–48]. It has been observed that the application of manure preserves the nutrients in soil, which are available for plant uptake, for a long period [49]. Therefore, in this study, SOC, soil TN and TP content indicated notable positive association with annual crop yield (Figure 2). Continuous manure input also improves the biological and physical properties of soil such as soil aeration, its water holding capacity and microbial community structure and biomass, which increase the crop yield. In contrast, NPK fertilization degrades the soil by decreasing soil pH and reducing the crop yield [46,50].

Subsequently, the application of manure has become a conventional practice, and the environmental impact of pig manure application has been previously studied [37,51,52]. In the present study, we found that the total As concentration was not increased compared to the control, but the available concentration of As was increased by increasing the manure input. However, its concentration did not cross the highest allowable level of As (>20 mg kg^{-1}) for agricultural soil (Table 1). Tang et al. [11] reported that no significant changes were observed after long-term pig manure application. Higher available As under NPK70%M treatment was due to the high soil pH, which induces the negative charges of the surface which can increase As desorption into the soil solution [7]. Moreover, an increase in the available concentration of As in soil was also associated with SOM, and long-term manure application acted as a storage for As in soil. In addition, during the decomposition of SOM, soil As was reintroduced into the soil solution [53]. Grain As concentration did not show significant changes among NPK, NPK30%M, NPK50%M and NPK70%M treatments (Figure 1b). However, compared to the control (CK) treatment, grain As concentration significantly increased under the fertilizer application, and grain As concentration crossed the allowable critical levels of As (<0.1 mg kg^{-1}), which can pose a serious risk to public health (Figure 1b). The addition of manure with NPK significantly decreased the bioaccumulation coefficient of As in comparison to the control and sole NPK (Figure 3). The lower BAC value by manure application might be associated with high SOC and pH because different species of As can make complexation with SOC under the high rate of manure input [7,11].

SOC, TN and TP content were positively correlated with available As concentration in the soil (Figure 4). Soil organic matter shows high affinity with As in soil [54]. In paddy soil, the adsorption of dissolved organic carbon on to hydroxides of iron via legend exchange significantly completes with As(III) and As(V) for active adsorption sites; therefore, dissolved organic carbon in soil can increase the As availability in paddy soil [55]. The positive relationships of TN and TP were associated with higher soil TN and TP content under manure application. In soil, inorganic P is mostly stable which is adsorbed on minerals in the soil, and a very small fraction of inorganic P exists in its ionic form; therefore, P only replaces the small proportion of As adsorbed on the mineral, unless soil is supplied with direct P application [56]. It has been observed that P and As compete with each other for binding sites which results in the release of soil-absorbed As(V) [57]. In the current study, the PLS test results show that the soil pH and SOC were the highly affecting factors on BAC under long-term fertilization, which explained the 75% and 17.6% variations, respectively (Figure 5). In paddy soil, high pH increases the desorption of As in solution and increases the availability of As for plant uptake [7], while pig manure mainly affects the bioaccumulation of As in paddy soil by changing the soil pH, SOC and nutrient content. Therefore, As concentration in manure should be monitored to reduce the bioaccumulation of As in rice grain while applying the manure to rice fields for high crop yield and food security.

5. Conclusions

In this study, we concluded that combined manure plus inorganic fertilization significantly improved the crop yield and soil nutrient availability. As in soil and rice grains was significantly correlated with soil properties, and soil pH was one of the most influencing factors on BAC, which explained 75.4% of total variations. Moreover, increasing the rate of pig manure in combination with inorganic fertilization also increased the plant-available As concentration in soil. However, the grain As concentration was significantly decreased by maximizing the rate of manure addition. Therefore, for sustainable and high crop production, it is necessary to monitor the concentration of As in manure so that it can assist in minimizing the transformation of As in the soil–plant system.

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