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ORIGINAL PAPER

Effects and Mechanism of Continuous Liming on Cadmium Immobilization and Uptake by Rice Grown on Acid Paddy Soils

Zhaobing Liu[1,2] · Ying Huang[1] · Xionghui Ji[2] · Yunhe Xie[2] · Jianwei Peng[1] · Mamdouh A. Eissa[3] · Ahmed E. Fahmy[4,5] · Salah F. Abou-Elwafa[6]

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Abstract

Lime application is the most effective agricultural practice for the reduction of cadmium (Cd) bioavailability in acid soils. This study was conducted to investigate the impact of continuous liming across five consecutive growing seasons on the remediation of Cd in acid paddy soils, as well as rice yield. Two rice cultivars, i.e., Zhuliangyou 819 and Xiangwanxian 12, were cultivated in Cd-contaminated paddy soil for five consecutive growing seasons from 2014 to 2018. The investigated lime levels were 0, 450, 900, 1350, 1800, 2250, 3000, and 3750 kg ha^{-1}. Lime application significantly increased rice yield, soil pH, exchangeable soil Ca^{2+}, and rice calcium (Ca) contents; besides, it reduced soil and rice Cd contents. The application of lime at the rate of 1350–2250 kg ha^{-1} significantly increased rice yield. Under continuous liming, rice yield obviously increased first and then decreased with the cumulative application of lime. The application of a cumulative lime amount of 18,000 kg ha^{-1} was identified as the critical transition point of soil pH, soil Cd, and rice Cd content. Application of lime up to or above 3000 kg ha^{-1} per season reduced Cd content in brown rice below 0.20 mg kg^{-1}. The results suggest that the reduction in effective Cd content might be a result of the combined action of exchangeable soil Ca^{2+} and soil pH rather than being a direct effect of Ca^{2+}. Therefore, acid Cd-contaminated paddy fields can realize the safe production of rice by the continuous application of an appropriate amount of lime.

Keywords Rice · Lime application · Cadmium · Heavy metals · Paddy soils

1 Introduction

Rice (*Oryza sativa* L.) is one of the most globally important food crops, especially in Asia and the Pacific region (Li et al. 2017). The continuously growing world population and the adverse impacts of climate changes necessitate carrying out research aimed at ensuring the sustainable production of such a staple crop and food security through agro-ecological managerial practices (El-Mahdy et al. 2018; Eissa and Negim 2018; Abou-Elwafa et al. 2019).

Acid soil is identified as the soil with a pH < 5.50 in its surface horizon (0–20 cm). About 30% of the global land area,

Zhaobing Liu and Ying Huang contributed equally to this work.

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✉ Jianwei Peng
314967900@qq.com

[1] College of Resources and Environment, Hunan Agricultural University, Changsha 410128, Hunan, China

[2] Ministry of Agriculture Key Laboratory of Agriculture Environment in Middle Reach Plain of Yangtze River/ Key Lab of Prevention, Control and Remediation of Soil Heavy Metal Pollution in Hunan Province, Hunan Academy of Agriculture Sciences, Changsha 410125, Hunan, China

[3] Departments of Soils and Water, Faculty of Agriculture, Assiut University, Assiut 71526, Egypt

[4] Haikou Experimental Stations, Chinese Academy of Tropical Agricultural Sciences (CATAS), Haikou, China

[5] Atomic Energy Authority, Nuclear Research Centre, Soil & Water Research Department, Abou-Zaabl 13759, Egypt

[6] Agronomy Departments, Faculty of Agriculture, Assiut University, Assiut 71526, Egypt

corresponding to 50% of the global arable area, has been identified as being affected by acidity (Dai et al. 2017). Soil acidity is a major yield-limiting factor that adversely affects crop productivity (Sumner and Noble 2003; Fageria and Nascente 2014). The adverse effect of soil acidity on crop productivity is mainly due to the fact that soil acidification greatly alters the physical and chemical properties of the soil as it affects the chemical and biological reactions in the soil, leading to substantial reduction in the availability of several essential nutrients (e.g., potassium, phosphorus, calcium and magnesium), while intensifying the toxicity level of other nutrients (e.g., Cd) (Sumner and Noble 2003). Soil acidification is mainly due to industrial pollution and agricultural practices, in addition to natural processes (Fageria and Nascente 2014; Holland et al. 2018; Eissa and Roshdy 2018).

Heavy metals are the most dangerous environmental pollutants and could be supplied to soils by agricultural management practices such as fertilizers application, pesticides and municipal wastes, atmospheric deposition, and sewage sludge (Abou-Elwafa et al. 2019). However, the uptake of heavy metals by plants depends to a great extent on their concentrations in soil and is controlled by their availability (Zaimoglu et al. 2009). Human and industrial activities have substantially increased heavy metals, Cd in particular, in the paddy soils (Wang et al. 2015). According to Chinese soil environmental quality limits (EQL), about 7.0% of cultivated soils, which are mainly distributed in the paddy regions, are contaminated with Cd (Liu et al. 2016). Consequently, almost 10% of brown rice production in China is contaminated with Cd (Li and Xu 2015; He et al. 2017). During the last two decades, heavy metal contamination of soils has become a serious problem in several geographic regions worldwide, including Asian, African, and Latin American countries, where rice production and consumption is rapidly growing (Hu et al. 2016). High concentrations of heavy metals are not causing toxicity problems only in plants but also in animals consuming plants and gradually deposited these metals leading to a bioaccumulation risk (Khanam et al. 2020). Cd was identified as a major toxic metal that can directly reach the food chain through crop uptake or can indirectly be transferred by animals (Grant 2011; Osman et al. 2015; Puga et al. 2015). The effects of Cd toxicity in humans and animals are mainly observed in growth inhibition, hypertension, and other diseases. In addition, Cd toxicity has adverse effects on the absorption of the enzyme system, fertility, and certain essential nutrients. It could also affect the sex of the fetus (Baba et al. 2013; Skroder et al. 2015).Therefore, the control and remediation of Cd-contaminated soil is extremely urgent to ensure food safety in southern China. Accumulation of heavy metals, Cd in particular, in rice grains can be decreased by decreasing their bioavailability and uptake by rice plants. Several approaches have been implemented to reclaim Cd-contaminated soils including phytoremediation, which uses hyperaccumulators to uptake Cd from soil, bioremediation, chemical amendments, agricultural practices, and engineering measures like replacement with clean soils, soil washing, and flushing. In spite of considering cost and efficiency, their use on large areas of agricultural soil cannot be promoted (Khanam et al. 2020; Li et al. 2020; Hirzel et al. 2019). The most effective and economically viable way of in situ remediation method for polluted agricultural soils is Cd immobilization with amendments. Many researches have proven the effect of lime, slags, red mud, and other amendment materials on the reduction of the effective content of Cd.

Among all the amendments, lime application is the most effective agricultural practice for improving crop productivity by (i) substantially reducing soil acidity by neutralizing the excessive hydrogen ions in the soil solution, (ii) enhancing the availability of essential nutrients and reducing the availability of toxic elements, (iii) directly supplying several essential elements for crop production (e.g., calcium (Ca^{2+}) and magnesium (Mg^{2+}) as constituent elements in lime materials, and (vi) indirectly affecting nutrients transmission and uptake by plants through affecting soil microbial activity (Fageria and Baligar 2008; Eissa et al. 2013; Cheng et al. 2013; Fageria and Nascente 2014; Goulding 2016; Kunhikrishnan et al. 2016; Youssef and Eissa 2017). Numerous reports on the application of lime for the remediation of heavy metal-contaminated soils have been published (Simón et al. 2010; Hale et al. 2012; Chen et al. 2016; Cui et al. 2016). In some previous studies, the application of lime significantly reduced Cd accumulation in rice grain below the Cd limiting standards (Bian et al. 2016; Li et al. 2016a; Xiao et al. 2017). In these cases, lime was applied to heavy metal-contaminated paddy soils to immobilize Cd (Hu et al. 2016). The application of lime to control the migration of heavy metals has many problems that should be considered and explored, especially the large increase in soil pH and the introduction of a large amount of calcium ions that will increase the risk of heavy metal dissolution in the soil after continuous application of lime, leading to the cumulative amount of absorption by plants.

In the current study, we hypothesize that continuous application of lime has a great effect on bioavailability and immobilization of Cd through increasing soil pH and increasing the availability of exchangeable soil Ca^{2+} via the antagonistic actions of Cd-Ca. Therefore, field experiments were conducted to investigate (i) the effect of lime application on improving rice yield in acid paddy soils, (ii) the impact of continuous liming on Cd and calcium availability in soil, and (iii) the effect of continuous liming on the Cd uptake and the interaction mechanism with calcium, and clarifying the related mechanism of lime-remediation in Cd-contaminated soils.

2 Material and Methods

2.1 Plant Material and Field Experiments

Two rice cultivars were used, i.e., the early-maturing cultivar, Zhuliangyou 819 (Zhu819), and the late-maturing cultivar, Xiangwanxian 12 (Xiang12). The field experiments were carried out for five consecutive growing seasons from 2014 to 2018 in Cd-contaminated paddy soils in Beishan Town, Changsha County, Hunan Province of China (E28°26′24″, N 113°03′29″), which has a subtropical monsoon climate with mild climate and abundant precipitation (average annual rainfall is 1171.6 mm). Soil Cd (Cd) pollution was caused by upstream chemical plants sewage at the end of the last century. The soil pH was 4.95, the organic matter was 49.3 g kg^{-1}, total nitrogen was 2.59 g kg^{-1}, alkali-hydrolytic nitrogen was 173.0 mg kg^{-1}, available-P was 35.2 mg kg^{-1}, available-K was 72.0 mg kg^{-1}, total-Cd was 1.02 mg kg^{-1}, available-Cd was 0.27 mg kg^{-1}, and exchangeable-Ca was 716.2 mg kg^{-1}.

Lime was applied in March and July every growing season. The locally produced slaked lime ($Ca(OH)_2$) which contains 0.46–0.88 mg kg^{-1} of Cd and 43.7–47.2% of CaO was added each year before cultivation. A basal dose of 600 kg ha^{-1} and 750 kg ha^{-1} of a compound fertilizer (NPK 22–5-13) with undetectable Cd content with topdressing doses of urea of 60 kg ha^{-1} and 75 kg ha^{-1} were applied to the early- and the late-maturing cultivars as follows: Zhuliangyou 819 and Xiangwanxian 12, respectively. After each application of lime, the soil was thoroughly homogenized; afterwards, the base fertilization dose was applied after 7 days after equilibrium, and then 2-week-old rice seedlings were transplanted. Other cultural managements followed locally recommended practices of rice cultivation. Early-maturing rice cultivar was harvested between the middle and late of July every year, while late-maturing rice cultivar was harvested in late October every year. Irrigation was performed using tap water where Cd had not been detected.

2.2 Experimental Design and Treatments

Experimental design followed the randomized complete block design (RCBD) in a split plot design with three replicates. Eight lime gradient dosage treatments, i.e., L0 (No lime), L450 (450 kg ha^1 lime), L900 (900 kg ha^{-1} lime), L1350 (1350 kg ha^{-1} lime), L1800 (1800 kg ha^{-1} lime), L2250 (2250 kg ha^{-1} lime), L3000 (3000 kg ha^{-1} lime), and L3750 (3750 kg ha^{-1} lime) were allocated to the main plots and the two cultivars to the subplots, with a plot area of 13 m^2 (5 × 2.6 m). The density of early rice was 4 × 6 in. and 5 × 6 in. for late rice. The experiment began in March 2014 and finished in October 2018. Each year, the lime was applied at the beginning of the planting season.

2.3 Sampling and Analyses

For basic analysis of the soil's physical and chemical properties, composite representative soil samples collected from 0 to 20-cm plowed layer of five sampling sites before the application of lime were air-dried and sieved through 20, 60, and 100 mesh sieves. During the growing season, soil and plant samples were collected once per season. For efficient determination of soil pH, exchangeable calcium, and Cd contents, soil samples were air-dried and sieved through a 20-mesh sieve. Five rice plant samples sampled randomly from each plot were washed with tap water, dried and ground, and then sieved using a 100-mesh sieve. Soil available Cd was extracted with NH_4OAc (Andrews et al. 1996) and exchangeable calcium was extracted with 1 mol L^{-1} NH_4OAc (Page et al. 1982). Soil pH (H_2O immersion, 2.5:1) was determined by acidity meter. The total Cd in the soil was digested with HNO_3-$HClO_4$-HF (5:1:2, V/V). After the sample was completely dehydrated, it was acid-treated to near dry. After adding a small amount of diluted nitric acid solution, it was transferred to known volume. The rice plant and brown rice Cd were treated with HNO_3-H_2O_2 (5:2, V/V) microwave digestion and quality was controlled by standard materials. The Cd content of all samples was measured by inductivity coupled plasma emission (ICP-MS (iCap-Q, Thermo Corporation, USA)), and the calcium content was determined by flame atomic absorption spectrometry. Sample replicates, reagent blanks, and standard reference materials were included in each batch of analysis to ensure analytical quality. The recovery of spiked standard for each batch ranged between 90 and 110%.

2.4 Statistical Analysis

Analysis of variance (ANOVA) and least significant difference (LSD) were performed using the software SPSS 17.0 package (SPSS, Chicago, IL, USA), and $P < 0.05$ was used to indicate statistical significance. The SigmaPlot 14 Software (Systat Software, San Jose, CA, USA) was implemented to perform linear regression and Pearson correlation coefficients (R) among measured traits. The regression model which resulted in the highest R^2 value was used in determining the relationships between cumulative lime application and the availability of Cd and Ca elements in the soil and the soil pH, and between the availability of either elements in the soil.

3 Results

3.1 Effect of Lime Application on Grain Yield

Significant differences in grain yield among either cultivar, growing seasons, or lime treatments were revealed by the analysis of variance (Fig. 1a; Supplementary Table 1). Generally, the application of a proper dose of lime in the acid Cd-contaminated rice fields increased rice grain yield. The application of doses of lime 1350, 1800, and 2250 kg ha^{-1} significantly increased rice grain yields. The grain yield of the early-maturing cultivar was significantly increased up to 20.9%, 18.8%, and 20.1% when doses of 1350, 1800, and 2250 kg ha^{-1} of lime, respectively, were applied in the 2014 growing season, as compared with the control treatment (L0) (Fig. 1a). Meanwhile, in the 2015, 2016, and 2018 growing seasons, the effect of lime treatment on rice grain yield did not reach the significance levels. In the 2017 growing season, the application of 1350 kg ha^{-1} of lime led to a highly significant increase (38.8%) in rice grain yield of the early-maturing cultivar. However, in the late-maturing rice cultivar, the application of a lime dose of 1800 kg ha^{-1} significantly increased rice grain yield by 28.5% in the 2016 growing season. However, in the 2017 growing season, the highest significant increase (20.6%) in grain yield resulted from the application of 2250 kg ha^{-1} of lime. In the 2018 growing season, the application of 1350 kg ha^{-1} of lime resulted in the significantly highest grain yield increment (19.2%). Meanwhile, the effect of lime treatment on grain yield in the 2014 and 2015 growing seasons was insignificant.

Linear regression analysis between cumulative lime application and rice grain yield across the five growing seasons fitted to the linear equation (Fig. 2A), which indicates that rice grain yield significantly increased in response to the cumulative application of lime, and then it decreased, indicating that the application of a proper amount of lime to the acid Cd-contaminated paddy soils can significantly increase rice grain yield. When the cumulative amount of lime reached about 28,250 kg ha^{-1}, the rice grain yield reached its maximum, but the effect decreased after exceeding that cumulative amount.

3.2 Effect of Lime Application on Soil pH

In general, soil pH increased in response to increasing lime application (Fig. 1b). Soil pH significantly increased when lime was applied at 1350 and 3750 kg ha^{-1} up to 0.56 and 1.15 pH units compared with L0 in the 2014 growing season. However, the effect of the application of a given amount of lime on increasing soil pH was declining from one season to another, e.g., the application of 1000 kg ha^{-1} of lime in the

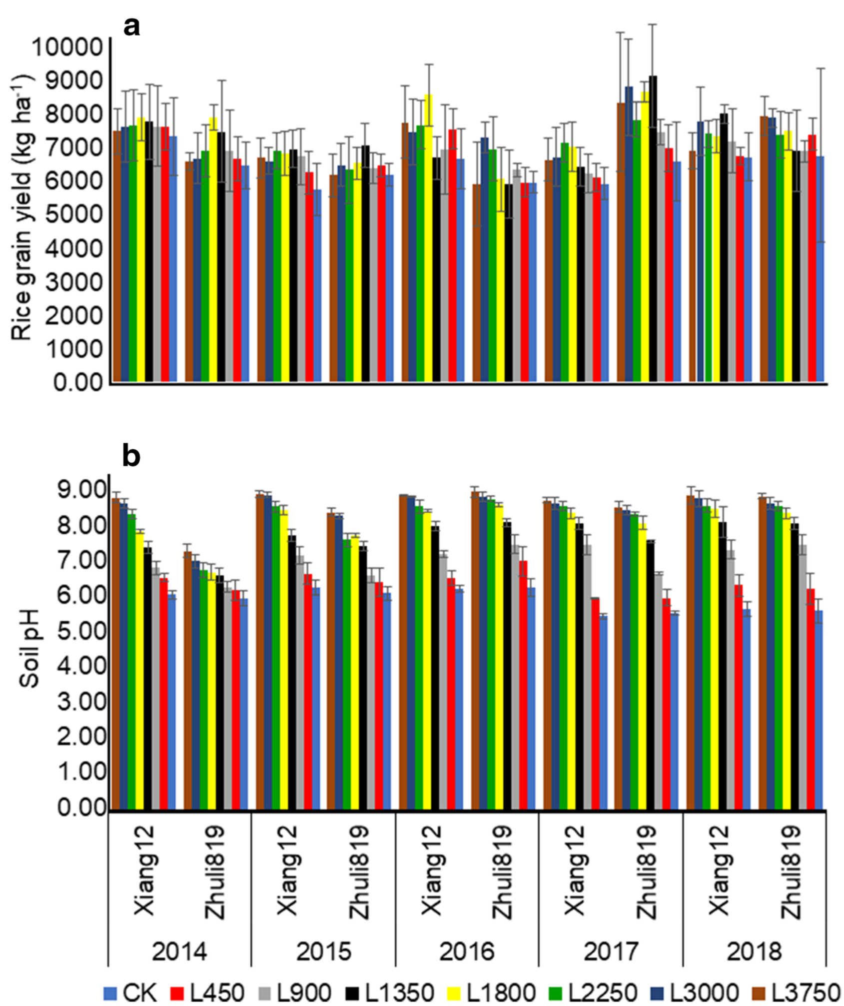

Fig. 1 Rice grain yield two rice cultivars, i.e., the early-maturity Zhuliangyou 819 (Zhuli 819) and the late-maturity Xiangwanxian 12 (Xiang 12), (**a**) and soil pH (**b**) as influenced by continuous lime application in five consecutive growing seasons (2014–2018) in acid paddy. The statistical analysis of the data is presented in Supplementary Table 1. CK, L450, L900, L1350, L1800, L2250, L3000, and L3750 indicate 0, 450, 900, 1350, 1800, 2250, 3000, and 3750 kg ha-1 of the cumulatively applied amount of lime

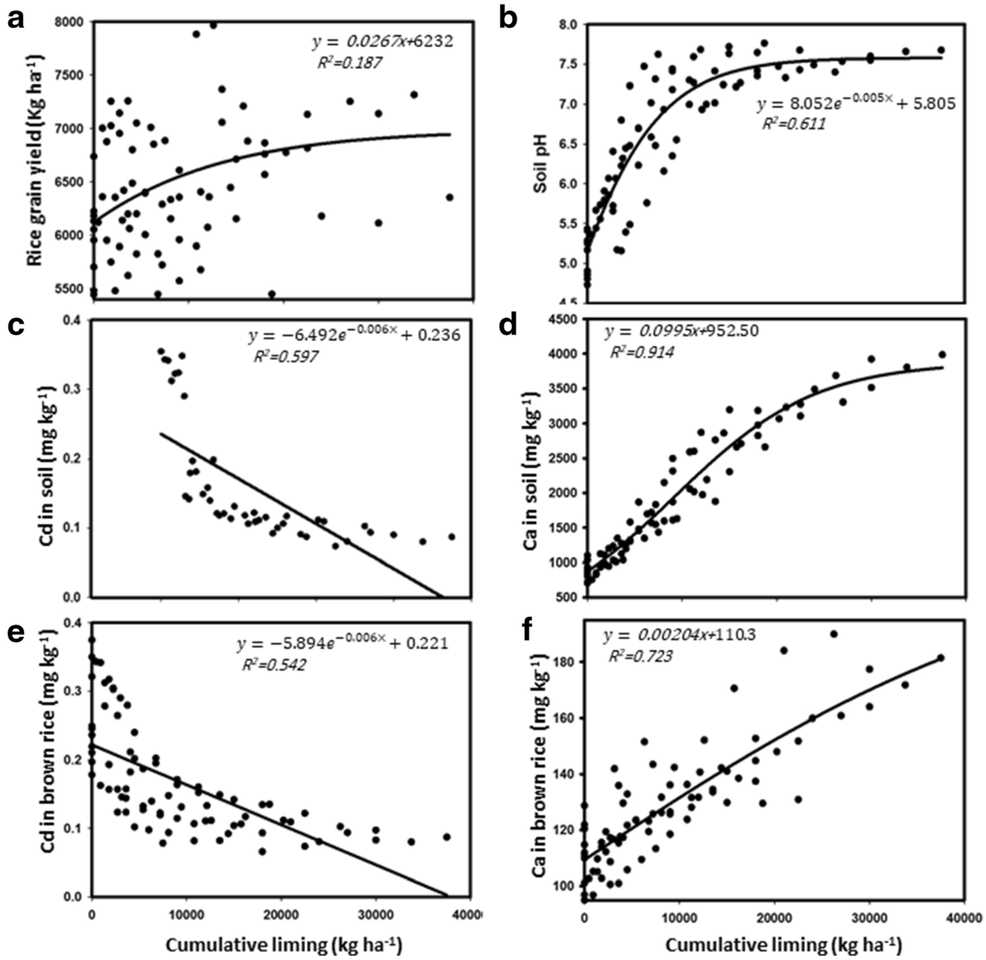

Fig. 2 Response of rice grain yield (**a**), soil pH (**b**), soil Cd content (**c**), soil Ca content (**d**), brown rice Cd content (**e**), brown rice Ca content (**f**) to continuous liming fitted by the linear-linear model across five consecutive growing seasons (2014–2018)

first growing season increased the soil pH by about 0.30 units, while in the fifth growing season, the application of the same amount of lime increased the soil pH by only 0.07 units (Table 1). Soil pH significantly correlated with the amount of applied lime and it rises with the increase in lime application. Linear regression indicates a single-peak curve relationship between the soil pH and the cumulative application of lime, and the cumulative effect of lime on the soil pH has gradually decreased (Fig. 2b). The regression analysis between the soil pH and the cumulative amount of lime corresponding to each treatment across the five growing seasons conform to the linear equation, and there was a significant variable leap characteristic in soil pH change. The soil pH rapidly increased in response to increasing the cumulative application of lime when the soil pH was less than 7.40 or the cumulatively applied amount of lime was less than 18,000 kg ha^{-1} (8100 kg ha^{-1} CaO). However, when the soil pH reached 7.40 or the cumulatively applied amount of lime reached about 18,000 kg ha^{-1} (8100 kg ha^{-1} CaO), the soil pH was almost stable.

Table 1 Correlation analysis between soil pH and cumulative amount of lime applied across five consecutive growing seasons (2014–2018)

Time	Fitting equation	R^2	P
Early-maturity rice cultivar_ 2014	$y = 3e^{-04x} + 5.220$	0.978	0.000
Late-maturity rice cultivar_ 2014	$y = 3e^{-04x} + 5.411$	0.950	0.000
Early-maturity rice cultivar_ 2015	$y = 2e^{-04x} + 5.396$	0.925	0.000
Late-maturity rice cultivar_ 2015	$y = 2e^{-04x} + 5.667$	0.896	0.000
Early-maturity rice cultivar_ 2016	$y = 1e^{-04x} + 5.894$	0.842	0.001
Late-maturity rice cultivar_ 2016	$y = 1e^{-04x} + 5.678$	0.862	0.001
Early-maturity rice cultivar_ 2017	$y = 1e^{-04x} + 5.155$	0.853	0.001
Late-maturity rice cultivar_ 2017	$y = 9e^{-05x} + 5.370$	0.754	0.005
Early-maturity rice cultivar_ 2018	$y = 8e^{-05x} + 5.469$	0.791	0.003
Late-maturity rice cultivar_ 2018	$y = 7e^{-05x} + 5.490$	0.805	0.003

3.3 Effect of Lime Application on Soil Available Cd

The available Cd content in the soil across all five growing seasons significantly decreased in response to increasing lime application rate (Fig. 3a; Supplementary Table 1). For instance, in the 2014 growing season, the available Cd content of soil reduced by 2.0% to 20.2% compared with L0 when the

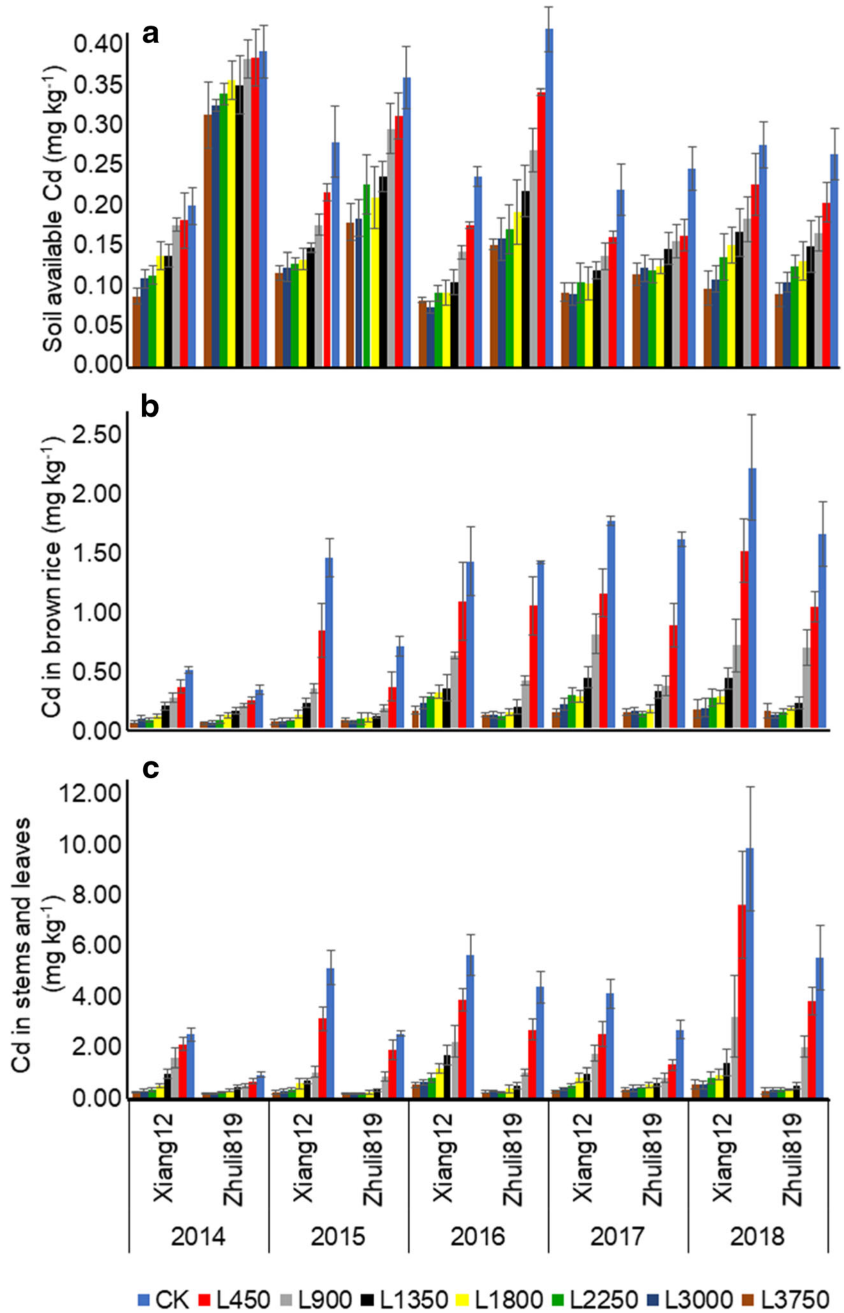

Fig. 3 Effect of continuous lime application on cadmium content in stems and leaves (**a**), in soil (**b**), and in brown rice (**c**) across five consecutive growing seasons (2014–2018) in acid paddy soils cultivated by two rice cultivars, i.e., the early-maturity Zhuliangyou 819 (Zhuli 819) and the late-maturity Xiangwanxian 12 (Xiang 12). The statistical analysis of the data is presented in Supplementary Table 1. CK, L450, L900, L1350, L1800, L2250, L3000, and L3750 indicate 0, 450, 900, 1350, 1800, 2250, 3000, and 3750 kg ha-1 of the cumulatively applied amount of lime

lime application rate was increased from 450 up to 3750 kg ha^{-1}, and the inhibition effect of Cd was obvious when the lime application rate reached 1350 kg ha^{-1}. Under continuous liming conditions, the application of an annual lime rate of 450 kg ha^{-1} significantly reduced the soil effective Cd content, and the greater the amount of applied lime, the lower the soil effective Cd content. The linear regression analysis between soil available Cd content and lime application rate showed a significant linear negative correlation ($R^2 = 0.597$, $P = 0.000$), while the linear regression analysis between soil available Cd content and the cumulative lime application rate across the five growing seasons was significantly fitted to the fourth-order equation (Fig. 2c). Increasing the cumulative application amount of lime up to less than 18,000 kg ha^{-1} (8100 kg ha^{-1} CaO) has rapidly decreased the soil available Cd content. However, when the cumulative application amount of lime reached 18,000 kg ha^{-1}, the decreasing rate in soil available Cd content was reduced and tended to be stable, and no reduction in the soil available Cd content occurred when the cumulative application amount of lime exceeded this level.

3.4 Effect of Lime Application on Soil Exchangeable Ca Content

Growing season, cultivars, and lime application rates exhibited significant effects on the soil exchangeable Ca content. The soil exchangeable Ca content significantly increased up to 32.4% and 44.1% in the 2014 growing season for the early-maturing and late-maturing cultivars, respectively, when lime application reached 1350 kg ha^{-1} (Fig. 4a; Supplementary Table 1). While the lime application rates of 1800 and 1350 kg ha^{-1} had significantly increased the soil exchangeable Ca content up to 131.1% and 68.8% for the early-maturing and late-maturing cultivars, respectively, in the 2015 growing season, in the 2016 growing season, the soil exchangeable Ca content significantly increased up to 70.2% and 42.3% as the lime application rate reached 900 and 1800 kg ha^{-1} for the early-maturing and late-maturing cultivars, respectively. The soil exchangeable Ca content significantly increased up to 81.6% and 81.7% for the early-maturing and late-maturing cultivars in the 2017 growing season, when the lime

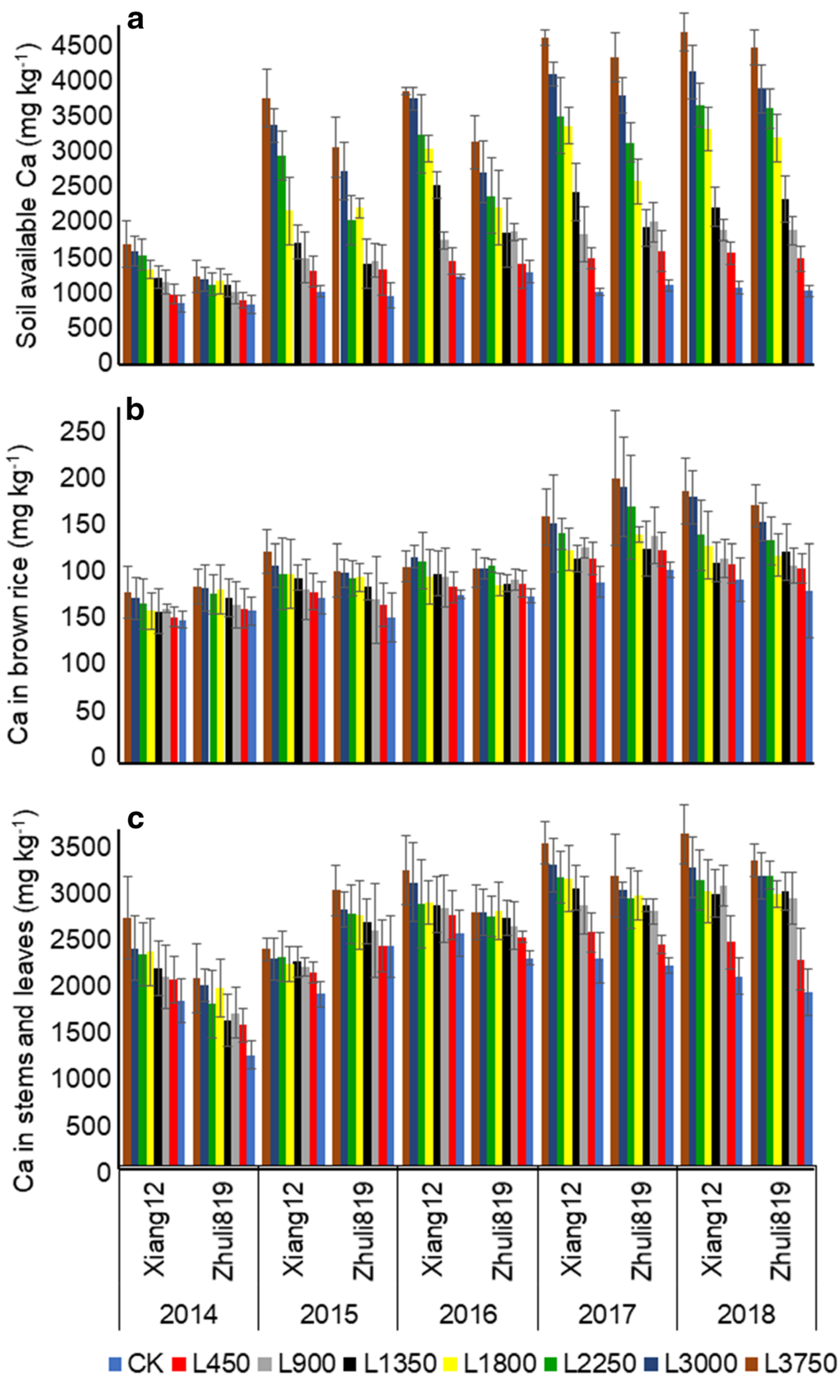

Fig. 4 Effect of continuous lime application on calcium content in stems and leaves (**a**), in soil (**b**), and in brown rice (**c**) across five consecutive growing seasons (2014–2018) in acid paddy soils cultivated by two rice cultivars, i.e., the early-maturity Zhuliangyou 819 (Zhuli 819) and the late-maturity Xiangwanxian 12 (Xiang 12). The statistical analysis of the data is presented in

application rate reached 900 kg ha^{-1}. In the 2018 growing season, the lime application rates of 900 and 450 kg ha^{-1} significantly increased the soil exchangeable Ca content up to 83.5% and 46.2% for the early-maturing and late-maturing cultivars, respectively. The linear regression analysis between the cumulative amount of lime applied and the soil exchangeable Ca content significantly fitted to the linear equation (Fig. 2d). A rapid characteristic increasing point in soil exchangeable Ca content was observed when the cumulative application amount of lime reached about 36,600 kg ha^{-1} (16,470 kg ha^{-1} CaO), where the amount of soil exchangeable Ca content greatly increased and the rate increment in soil exchangeable Ca content decreased.

3.5 Effect of Lime Application on Rice Cd Content

The results showed that the application of the proper amount of lime to acid paddy soil significantly inhibited the absorption and accumulation of Cd in rice. Cd content in brown rice, stem, and leaves decreased in response to increase in the rate of lime application (Fig. 3b, c; Supplementary Table 1). In all five growing seasons, the application of 450 kg ha^{-1} of lime either to the early- or the late-maturing cultivars significantly reduced Cd content in brown rice up to 24.0–49.4%. When lime was applied at the rate of 3750 kg ha^{-1}, the reduction in Cd content in brown rice ranged between 88.9 and 93.0%. Under continuous application of lime, Cd content in brown rice reduced to 0.2 mg kg^{-1} when lime was applied up to or above 3000 kg ha^{-1} per season (1350 kg ha^{-1} CaO). The changes in the Cd content in rice stems, leaves, and brown rice were basically similar. Compared with L0, in all five growing seasons, the application of 450 kg ha^{-1} of lime either to the early- or late-maturing cultivars significantly decreased the Cd content in the rice stems and leaves up to 15.0–51.6%. However, when the rate of lime application reached 3750 kg ha^{-1}, the Cd content of rice stems and leaves across all five growing seasons ranged between 82.0 and 95.7% lower than that of L0 (Fig. 3c).

The linear regression analysis between cumulative lime application and Cd content in brown rice across all five growing seasons was significantly fitted to the linear equation (Fig. 2e). The Cd content in brown rice also showed obvious climatic characteristics, that is, the Cd content of brown rice rapidly increased as the cumulative applied lime was below 18,000 kg ha^{-1} (8100 kg ha^{-1} CaO). Meanwhile, when the cumulative application amount of lime reached 18,000 kg ha^{-1} (8100 kg ha^{-1} CaO), the reduction rate in brown rice Cd content obviously decreased and tended to be stable as the cumulative application amount of lime exceeded 18,000 kg ha^{-1}.

3.6 Effect of Lime Application on Rice Calcium Content

Analysis of variance revealed that the application rates of lime and growing seasons exhibited significant effects on brown rice Ca content. The calcium content of brown rice was significantly increased in response to the increase in the application rate of lime (Fig. 4b; Supplementary Table 1). The Ca content of brown rice for the early- and late-maturing cultivars in the 2014 growing season increased as the rate of lime application increased; however, the differences were insignificant. In the 2015 growing season, the application of 1800 kg ha^{-1} of lime for the early and late-maturing cultivars significantly increased Ca content in brown rice by 27.4–32.2% compared with L0. The application of 3750 kg ha^{-1} of lime significantly increased Ca content in brown rice up to 28.3%, while the application of 2250 kg ha^{-1} of lime significantly increased Ca content in brown rice by 16.5–18.4%. However, only the application of 3000 kg ha^{-1} of lime led to a significant increase in the Ca content of brown rice up to 22.8–47.6% for the early- and late-maturity cultivars in the 2016 and 2017 growing seasons. The Ca content of brown rice significantly increased up to 24.6–49.8% in the 2018 growing season with the lime application rate of 2250 kg ha^{-1}. Linear regression analysis between the cumulative lime applied and Ca content in brown rice across all five growing seasons significantly fitted to the linear equation (Fig. 2f). The Ca content in brown rice rapidly increased as the cumulative amount of lime applied was below 120,000 kg ha^{-1} (54,000 kg ha^{-1} CaO). Meanwhile, when the cumulative amount of applied lime exceeded this level, the Ca content transition point was delayed.

The calcium content in rice stems and leaves significantly increased in response to increasing lime application rate, and the overall trend was similar to that in brown rice (Fig. 4c). There were also significant differences among different growing seasons. The application of 1800 kg ha^{-1} lime significantly increased the Ca content in rice stems and leaves in the 2014 growing season for the early-maturing cultivar. Meanwhile, the application of 3750 kg ha^{-1} lime resulted in the highest Ca content in rice stems and leaves for the late-maturing cultivar in 2014 and the early-maturing cultivar in the 2015 growing seasons. For the late maturity cultivar in 2015 growing season, the application of 900 kg ha^{-1} lime significantly increased Ca content in rice stems and leaves up to 16.0–26.5%. The application of 1350 and 3750 kg ha^{-1} lime for the early- and late-maturing cultivars, respectively, resulted in the significantly highest Ca contents in rice stems and leaves. In 2017 and 2018 growing seasons, the Ca content of rice stems and leaves was significantly increased when lime was applied at the rate of 900 kg ha^{-1} (Fig. 4c).

4 Discussion

Rice production is greatly affected by the environmental and socioeconomic factors that impact rice performance and production. There is a drastic decline in the growth rate and yield of rice, as land resources and water for its production are becoming scarce. In order to improve rice productivity, ensure food security for rice consumers, protect the environment, and combat poverty, great efforts have to be made towards attaining sustainable production increases. Therefore, the availability of information about technologies for designing regional programs to enhance and sustain rice production for durable food security and environmental protection are needed. The present study was conducted to evaluate the effect of cumulative lime application for five consecutive growing seasons on yield performance of two rice cultivars grown in acid Cd-contaminated paddy soils and its impact on the translocation of Cd from the soil to the plants to ensure sustainable, eco-friendly, and healthy rice production in such soils. Besides, understanding rice response to lime application in acid paddy soils is of great importance for attaining yield potential to meet the dynamic of population development. It has been widely reported that lime application in acid soils greatly improves crop productivity directly through enhancing soil physical, chemical, and biological properties, which ultimately increase the availability and mobility of plant essential nutrients (Chan and Heenan 1998; Bolan et al. 2003; Jaskulska et al. 2014; Al-Sayed et al. 2020; Rekaby et al. 2020). Besides, the increase in rice grain yield in acid soils in response to liming was mainly ascribed to the increase in soil pH which drastically decreases aluminum toxicity and promoted the absorption of Ca, Mg, P, and K (Rahman et al. 2005; Goswami et al. 2008; Previna and Baskar 2012; Fageria and Nascente 2014; Crusciol et al. 2016; Zeng et al. 2017). The effect of liming is mainly dependent on the duration of liming which involves a great discrepancy between different environmental conditions and managerial practices over time. However, continuous or excessive application of lime causes imbalance of Ca and Mg in the soil and inhibits Mg absorption by plants and could be detrimental to crop growth, leading to a reduction in the grain yield Carran 1991; Yi et al. 2006). Here, the results show that the application of a proper amount of lime in acid soil significantly increases rice grain yield, and the continuous application of large amounts of lime leads to a decline in the increment in grain yield, indicating that excessive lime application is not conducive to rice growth as it ultimately impacts rice grain yield.

Neutralizing soil acidity by reducing excessive hydrogen ions from the soil solution is the primary purpose of lime application (Bolan et al. 2003; Pagani and Mallarino 2012). The results indicate that cumulative application of lime is the most important factor in increasing soil pH. A significant linear relationship was observed between the response ratio of soil pH and the cumulative application of lime, and the model explains more than 61.1% of the variations (Fig. 2b). However, the complexity of the R^2 value, a factor which affects soil pH, was noticeable. Regardless of experimental and environmental conditions, a cumulative lime amount of 18,000 kg ha^{-1} or a pH value of 7.40 was the most effective threshold of cumulative amount of lime applied. The data reveal that a cumulative amount of 18,000 kg ha^{-1} of lime is the most effective threshold for increasing soil pH to 7.40 (Fig. 2b). The effect of lime application tends to be higher in the first growing season when the initial soil pH was low, then it declined in the following growing seasons as the soil pH increased (consistent with (Merida-Garcia et al. 2019)). These results should be useful for farmers worldwide as they will help to reduce the cost of lime application. Furthermore, the effect of lime application on soil pH depends to a great extent on some soil physical and chemical properties that affect soil buffering capacity such as cation exchange capacity, soil dehydrogenase activity, soil texture, and soil bacterial diversity (Bolan et al. 2003; Duan et al. 2020; Filipović et al. 2020). The application of lime to the acid Cd-contaminated paddy soils greatly improved soil pH, and it peaked at the application of 3750 kg ha^{-1} of lime (Fig. 1b).

Soil pH is extremely important for the activity and adsorption of toxic metals in the soil (Dinesh and Kumar 2006; Adhikari and Singh 2008; Choppala et al. 2013; Azouzi et al. 2015). Therefore, since most of these toxic metals are less soluble under alkaline conditions, increasing soil pH has become a common procedure for repairing heavy metal-contaminated soils (Adriano 2001; Eissa et al. 2016; Eissa 2019; Almaroai and Eissa 2020). However, if the soil pH is too high it would cause dissolution of heavy metals. Although the adsorption of heavy metals increases in response to the increase in soil pH within a certain range, their mobility is reduced in strong alkaline conditions due to the formation of hydroxy complexes, which ultimately leads to metals precipitation (Heasman et al. 1997; Wang et al. 2006; Loganathan et al. 2012; Sun and Xu 2013; Rees et al. 2015). Our data show that under continuous liming conditions, due to the cumulative effect of lime, its application at an annual rate of 450 kg ha^{-1} significantly reduces the soil effective Cd content. Besides, the cumulative amount of lime applied (18,000 kg ha^{-1}), corresponding to soil pH transition point (7.40), and this is basically the same amount as the soil effective Cd critical point. In addition to neutralizing excessive acidic proton ions and cations in the soil (e.g., Cd^{2+}), lime application supplies basic cations to the root zone (e.g., Ca^{2+}; Fageria and Baligar 2008). Our data show that lime application decreases soil available Cd content while increasing the exchangeable Ca content. The application of an annual lime rate of 450 kg ha^{-1} significantly reduced the soil effective Cd. The data demonstrate that a cumulative application of 18,000 kg ha^{-1} of lime is a critical threshold for reducing the soil Cd content, while the cumulative applied amount of 36,600 kg ha^{-1} of lime was considered a critical point for the exchangeable soil Ca content. Increasing Ca availability in response to reducing Cd availability was anticipated due to the decrease in the antagonistic actions of Cd-Ca as liming reduces exchangeable or available Cd^{2+} (Wu et al. 1992). Besides, the effect of increasing soil pH, caused by the continuous application of lime, on Cd availability, may be much greater than that of Ca^{2+}, as evidenced by Figs. 5 a and b. Therefore, in this study, the reduction in Cd effectiveness might be a result of the combined action of Ca^{2+} and pH rather than a direct effect of Ca^{2+}.

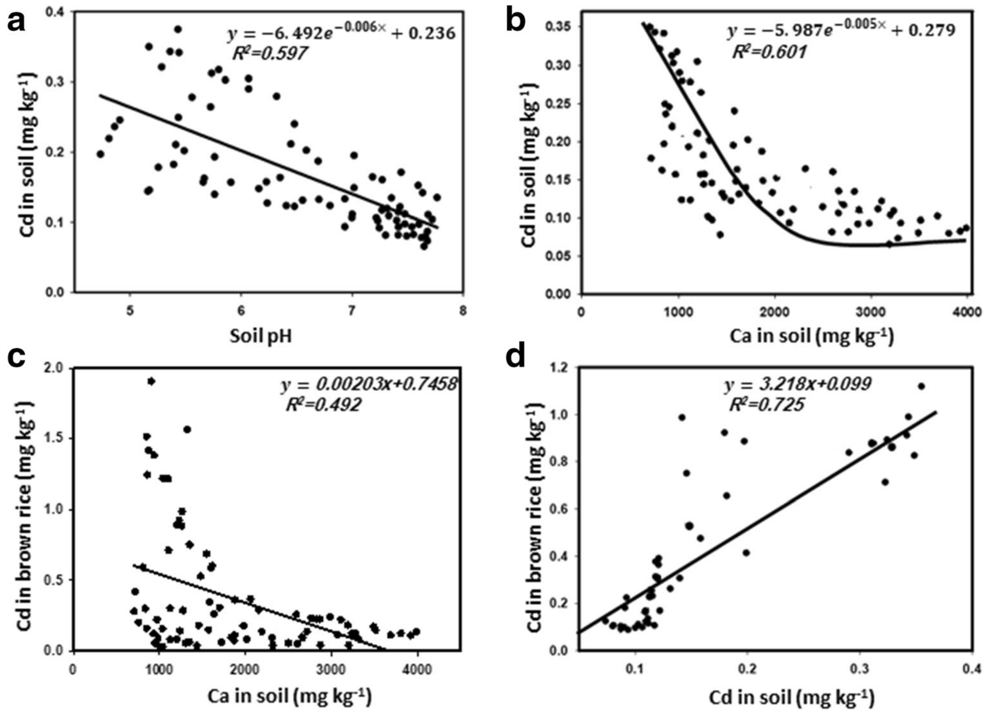

Fig. 5 Soil available Cd content response to soil pH (**a**), soil available Cd content response to soil available Ca content (**b**), brown rice Cd content response to soil available Ca content (**c**), brown rice Cd content response to soil available Cd content (**d**) fitted by the linear-linear model across five consecutive growing seasons (2014–2018)

As calcium and Cd compete for absorption by plant roots, the higher calcium content in lime crucially reduces the absorption of accumulated Cd by crops. It was believed that the competition between Ca^{2+} and Cd^{2+} was one of the important reasons for the inhibition of Cd migration and therefore significantly increased calcium content in plants and reduced the accumulation of Cd in plants (Zhou et al. 2001; Roosens et al. 2003; Österås and Greger 2006; Hong et al. 2009; Wang et al. 2013; Li et al. 2016b). In this study, under continuous application of lime, the Cd content of brown rice reduced to 0.20 mg kg^{-1}, when lime was applied up to or above 3000 kg ha^{-1} per season (Fig. 3b) which meets the requirements of the national food safety standard of the People's Republic of China (GB2762–2017). Cd content in brown rice was significantly adversely correlated with soil exchangeable Ca content (Fig. 5c), which confirmed the important effect of calcium on the absorption and accumulation of Cd in rice. Moreover, the Cd content in brown rice rapidly decreased as the exchangeable Ca content in soil increased within a certain range, after which the reduction rate of the Cd content of brown rice decreased. Therefore, the available soil Cd content under the experimental conditions cannot be considered as the only factor for characterizing the absorption and accumulation of Cd in rice, because the Ca^{2+} effect was obvious after the continuous application of lime that can affect not only the adsorption and desorption of Cd^{2+}, and the effectiveness of Cd, but also can compete for adsorption sites in rice roots, which in turn affect the absorption of Cd by rice (Fig. 5d). The results further revealed that about 59.1% of the Cd content in brown rice was related to soil exchangeable Ca content, soil available Cd content, and soil pH.

5 Conclusions

Adoption of an effective agronomic approach for the production of rice with low cadmium (Cd) accumulation could be a realistic option that would allow rice cultivation in paddy soils with high bioavailability of such a toxic element. Continuous liming is a powerful and cost-effective agronomic approach that could help in mitigating the bioavailability, immobilization, and accumulation of Cd in paddy soil as well as plant parts. The results further suggest that the reduction in effective Cd content might be a result of the combined action of calcium (Ca^{2+}) and soil pH rather than being a direct effect of Ca^{2+}. Therefore, safe production of rice can be achieved in Cd-contaminated acid paddy field through continuous application of an appropriate amount of lime. Continuous application of lime up to or above 3000 kg ha^{-1} per season reduces the Cd content in brown rice below 0.20 mg kg^{-1}, which meets the requirements of the national food safety standard of the People's Republic of China.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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