Effects of Rice Straw, Rice Straw Ash, and Bone Charcoal on Uptake and Accumulation of Rare Earth Elements in Rice Plants

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Pot experiments were conducted to study the effects of rice straw (RS), rice straw ash (RSA), and bone charcoal (BC) on the bioavailability of 15 rare earth elements (REEs) in soil and the absorption and accumulation of REES by rice. Adding RSA and BC to REE-contaminated soil remarkably increased the biomass and yield of rice, and the addition of RS remarkably inhibited the growth of rice. Compared with the control check (CK), the total REE concentration in the soil solution at the tillering stage, heading stage, and maturity stage was significantly increased by adding RS, and the total REE concentration in the soil solution was remarkably decreased by adding RSA and BC. The concentration of 15 REEs in the roots, shoots of rice, and brown rice were remarkably decreased *via* RSA addition. The concentration of total REEs in rice roots, shoots, and grains decreased 79.1%, 76%, and 18.3%, respectively, and the concentration of total REEs in the roots and shoots of rice decreased 19.9% and 67.2%, respectively *via* RSA addition. However, there was no noticeable effect on the concentration of total REEs in brown rice. So BC and RSA are suitable to be added to REE-contaminated soil, but RS is not.

Keywords: Rice straw; Rice straw ash; Bone charcoal; Rare earth elements; Rice plants

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INTRODUCTION

Rare earth element (REE) is the general term used for the members of the third subgroup B elements (IIIB) family in the periodic table, *i.e.*, scandium, yttrium, and the lanthanides. The lanthides are the 15 chemical elements with atomic numbers 57 through 71 (Cornell *et al.* 1993; Zhao and Wilkinson 2015). The properties of REEs are very similar in three ways: (1) the atomic structure is similar; (2) the ionic radius is similar; and (3) the common valence is +3, which allows stable coordination compounds to form easily. Thus, they are closely symbiotic in nature (Hu *et al*. 2006; Aquino *et al*. 2009). The most common grouping method for REEs is dichotomy, namely by light REEs and heavy REEs. Light REEs are also known as cerium REEs and include La through Eu on the periodic table. Heavy REEs, also known as yttrium rare earth, include Gd through Lu, Y, and Sc (Wyttenbach *et al*. 1998; Tyler 2004; Wiche *et al*. 2016). China, mainly South China with South Jiangxi as its center, is rich in rare earth resources (especially ionic-type rare earth resources), accounting for 90% of the global supply of rare earth resources. During the process of the exploitation and smelting of rare earth resources, the

REE enrichment of soil, water, and the crops surrounding the mining areas becomes higher due to the lack of supervision, illegal exploitation, outdated technology, and emission of the industrial “Three Wastes” (waste gas; waste water; industrial residue) (Pan and Zhu 2003; Jin and Huang 2013, 2014; Wiche *et al*. 2016; Suja *et al*. 2017; Zhuang *et al.* 2017). Rare earths are not necessary nutrients for the growth of plants and animals. Small amounts of REEs can promote growth, yet large amounts can inhibit growth and even lead to poisoning (Jin *et al*. 2016; Wiche *et al*. 2016; Gravina *et al*. 2018; Zheng *et al*. 2018). With the extensive use of REEs in many industries, the accumulation of rare earths in the environment is steadily getting worse, and REEs enter the human body through the consumption of food (Wang *et al*. 2017; Kyra *et al*. 2017). It has been reported that the concentrations of REEs in the hair, blood, bone, brain, and liver of residents of mining areas are remarkably higher than those in non-mining areas (Wei *et al*. 2013; Li *et al*. 2014). This affects the health of the mining area residents, as shown *via* abnormal biochemical indices of the blood, abnormal functioning of the liver, and lower IQ of children living in mining areas. High concentrations of REEs also adversely affect functions of the reproductive and immune systems (Zhang *et al*. 2000; Li *et al*. 2014; Liu *et al*. 2015). However, few studies have been done on how to reduce the uptake of REEs into the food chain from the soil.

South China (with South Jiangxi as its center) is an important global area for ionic rare earth distribution. The main soil in South Jiangxi is red soil, which is acidic and lacks organic manure and effective phosphate. The area is also an important grain-producing area, as hundreds of millions of tons of rice straw are produced annually. Rice straw is composed of large amounts of organic matter and inorganic matter. The organic matter is composed of cellulose and soluble organic matter. The silicate concentration of inorganic substances is higher than 12% (Ma *et al*. 2002). To avoid environmental pollution and the waste of resources, rural rice straw is generally returned to the field and used comprehensively. The return of rice straw to the field can increase the soil organic manure and improve soil properties and soil nutrients. There are two main forms of comprehensive utilization of rice straw: electricity generation *via* gasification and rice straw burning. The burning of rice straw forms a large amount of rice straw ash. The RSA is alkaline, contains potassium oxide, and, of particular note, comprises more than 70% silica (Ma *et al*. 2002). The addition of RSA to red soil can improve the soil properties and nutrient contents.

To date, many studies have examined the effects of dissolved organic matter (DOM), phosphorated material, and silicate on the form and availability of heavy metals in the soil for China and the rest of the world. Returning rice straw to fields can increase soil organic matter and form DOM. Dissolved organic matter is a negatively charged colloid that competes with heavy metals in soil. It can promote the dissolution and desorption of heavy metals in the soil and become a "carrier" in heavy metal migration and activation, thus affecting the bio-availability of heavy metals in the environment (Zhu *et al.* 2014, 2016; Rikta *et al.* 2018). Xu and Yuan found that as the DOM concentration increased, the concentration of water-soluble heavy metals in the soil gradually increased (Xu and Yuan 2009). Jin *et al*. (2016) found that the formation of DOM in soil resulted in a significant increase in the concentration of water-soluble REEs. In contrast, DOM has the ability to form complexes with heavy metals because it has a large number of functional groups, such as carboxyl, hydroxyl, and carbonyl groups. Organic matter can also form complexes with heavy metals by using Fe or S as a bridge directly or indirectly, thus reducing the mobility of heavy metals in soil (Perroeenet *et al.* 2000; Williams *et al.* 2011). The DOM can affect the bio-availability of heavy metals from two distinct aspects. Many studies have shown that siliceous ions react with heavy metals to form silicic acid precipitates when silicon-containing materials are applied to soil. Silicon-containing materials can increase the pH of soil, enhance the soil adsorption capacity, adsorb heavy metal ions or complexes with opposite charges in soil, and reduce the activity of heavy metals (Zhao *et al*. 2014; Rizwan *et al*. 2016a; Gao *et al*. 2018; Talitha *et al*. 2018). Silicon is an important component of cell walls. The silicon present in cell walls binds to Cd, attaching it to cell walls. Adding silicon to soil can enhance the cell wall retention of Cd in soil organisms (Liu *et al*. 2013; Duan *et al*. 2016). After bone charcoal is added to soil, the phosphoric acid ions in plant roots generated *via* the acidic soil can form a refractory metal phosphate with the REE ions. Bone charcoal is alkaline and has alkali ions, a large surface area, and abundant functional groups that contribute to metal complexation (Rinklebe *et al*. 2016; Rizwan *et al*. 2016b). The effects of bone charcoal when added to soil include the immobilization of heavy metals, modification of soil pH, and improvement in the physical and biological properties of soil (Rizwan *et al*. 2016b). However, it is not known whether the effects of soil organic matter, silicon-containing materials, and phosphorus-containing materials on the bio-availability of heavy metals fully reflect the impacts on the bio-availability of REEs. Hence, it is necessary to scientifically evaluate the effects of organic matter, silicon-containing materials, and phosphorus-containing materials on the yield, quality, and ecological environment of rice fields in rare earth mining areas. Furthermore, it is necessary to explore the rational restoration methods of rare-earth-contaminated soil, and provide guidance for the theory and practice of food safety, ecological environment protection, and the rational utilization of straw in rare earth mining areas.

EXPERIMENTAL

Materials

The test soil was collected from the rare-earth mine in the town of Jiading, Xinfeng County, Ganzhou City, Jiangxi Province, China. The soil sampling occurred as follows: the top 20 cm of soil was collected, dried naturally, removed of stones and plant residue, crushed, passed through a 100-mesh sieve, and stored until use. The physical and chemical properties of the soil and REE concentrations are shown in Table 1. The test rice straw (RS) was collected from the Xinzhou District of Jiangxi Province, China, which does not suffer rare earth pollution, and crushed into powder by a powder machine (Shanghai Precision Science Instrument Co., Ltd., Shanghai, China). The rice straw ash (RSA) was purchased from the Longshui Plant Straw Processing Plant in Ganyu County (Lianyungang, China). The bone charcoal (BC) was bought from the Tengzhou Chemical Plant in Shandong Province (Zaozhuang, China). The physical and chemical properties of the RS, RSA, and BC and their concentrations of REEs are shown in Table 1.

The rice variety was Jin Qian 47, and the rice seed was purchased from Jinhua Agricultural Science Institute of Zhejiang Province (Jinhua, China). The seeds were sterilized in a 10% H_2O_2 solution for 10 min, then washed with water several times. The seeds were planted in the test soil (free from rare earth pollution) for seedling raising.

Treatments

Pot experiments were conducted to study the effects of different soil amendments on the absorption and accumulation of REEs in rice. Four treatments were set: control group without modifier addition (CK), 2.5% rice straw (RS), 2.5% rice straw ash (RSA), and 2.5% bone charcoal (BC), with each treatment performed in triplicate.

Table 1. Physicochemical Properties and REEs Concentrations of Soil and Amendments

Items	Soil	RS	RSA	BC
pH	5.04	5.89	11.95	9.23
CEC (cmol.kg^{-1})	7.50	-	-	-
Organic matter (g.kg^{-1})	2.28	-	-	-
Total N (g.kg^{-1})	1.13	-	-	-
Total C (g.kg^{-1})	11.58	-	-	-
Total S (g.kg^{-1})	0.47	-	-	-
Soil particle size- Clay (%)	21.43	-	-	-
Silt (%) Sand (%)	68.53 10.05	- -	- -	- -
La (mg.kg^{-1})	145.09	0.6	1.27	2.37
Ce (mg.kg^{-1})	177.65	0.51	1.06	1.34
Pr (mg.kg^{-1})	40.29	0.17	0.3	0.41
Nd (mg.kg^{-1})	92.82	0.46	0.95	2.19
Sm (mg.kg^{-1})	18.49	0.13	0.22	0.36
Eu (mg.kg^{-1})	4.16	0.08	0.09	0.13
Gd (mg.kg^{-1})	9.58	0.13	0.21	0.28
Tb (mg.kg^{-1})	2.88	0.05	0.06	0.09
Dy (mg.kg^{-1})	6.39	0.08	0.14	0.21
Y (mg.kg^{-1})	52.47	0.29	0.56	1.16
Ho (mg.kg^{-1})	3.07	0.04	0.05	0.07
Er (mg.kg^{-1})	8.54	0.06	0.1	0.04
Tm (mg.kg^{-1})	1.20	0.03	0.04	0.02
Yb (mg.kg^{-1})	7.56	0.05	0.09	0.01
Lu (mg.kg^{-1})	1.07	0.03	0.03	0.002
Total REEs (mg.kg^{-1})	571.26	2.7	5.15	8.66

Notes: RS refers to rice straw, RSA refers to rice straw ash (RSA), and BC refers to bone charcoal.

In the experiment, a plastic basin with a height of 26 cm and a diameter of 24 cm was used, to which 5 kg of soil was added. To grow the rice plants, 5.4 g of urea (containing 46.6% N) and 0.6 g of potassium chloride (containing 62.9% K_2O) were used as fertilizer. According to the experimental design, the additives are added into the tested soil and stirred thoroughly until the additives are evenly distributed in the soil. An even mixture of soil amendments and the test soil was added into each plastic basin. The water balance was maintained for two weeks, and the water level was maintained at approximately 3 cm.

After three weeks of growth, the rice seedlings were transplanted to culture pots, with two seedlings per pot. After the transplantation, a soil solution extractor was embedded in each culture pot, deep in the rhizosphere of the rice, and used to collect the soil solution samples. The pot experiment was conducted in a greenhouse. The growth conditions of rice were cycles of light at 28 ℃ for 14 h and darkness at 20 ℃ for 10 h, luminous intensity of 260 to 350 $\text{mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$, and relative humidity of 60% to 70%. To ensure the uniformity of light and heat received per pot of rice, the position of each pot of rice was randomly changed every three days. During the entire test period, the soil in the basin remained flooded. The soil solution was collected at the tillering, heading, and maturity stages of rice and then filtered with a 0.45-m filter membrane.

After 1% nitric acid acidification, the solution was preserved in a refrigerator at 4 ℃ prior to testing. After the rice ripened, it was harvested. First, the rice was dropped manually from the rice panicle and loaded into a net bag. Then, the shoots and roots of the rice were harvested separately and rinsed with tap water first followed by three rinses with deionized water.

Water-absorbent paper was used to remove any leftover water by drying it to a constant weight in a 70 ℃ oven. The harvested rice was placed in an indoor area with ventilation and dried naturally; then, the husk was removed by a sheller and the grains collected. A stainless-steel grinder was used to grind the grains, shoots, and roots into powder, which was then stored at room temperature until analysis.

Table 2. Effects of Different Amendments on the Concentrations of Rare Earth Elements in Different Parts of Rice

Part	Materials	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	∑REE
Grain	CK	23.58b	66.64b	98.56b	77.06b	80.6b	70.26b	63.88b	45.8b	39.93b	44.88b	27.54b	40.69b	29.19b	32.27b	23.01b	763.89b
	RS	20.94a	53.78a	88.13a	76.02b	81.24b	67.22b	63.44b	42.85b	40.12b	44.49b	28.46b	41.82b	29.79b	35.88b	24.40b	738.58b
	RSA	23.53b	61.26b	95.96b	76.56b	82.86b	68.1b	65.16b	44.5b	39.05b	43.10b	27.45b	39.94b	29.14b	32.71b	23.01b	752.33b
	BC	18.38c	52.78a	79.12c	62.17a	68.63a	55.26a	52.62a	38.81a	32.61a	36.27a	22.79a	32.79a	24.27a	28.56a	19.31a	624.37a
Shoot	CK	1.12c	2.83b	2.35b	0.56b	1.96b	0.39b	0.14b	0.32c	0.08c	0.24c	0.07b	0.14c	0.04bc	0.11c	0.04b	10.39b
	RS	0.71b	1.65a	1.42b	0.38a	1.22a	0.27a	0.11a	0.30bc	0.06b	0.16b	0.05a	0.10b	0.04bc	0.09bc	0.03b	6.58a
	RSA	0.31b	0.83a	0.72a	0.21a	0.57a	0.16a	0.11a	0.13ab	0.05ab	0.10ab	0.04ac	0.07b	0.03ab	0.05ab	0.03ab	3.41a
	BC	0.25a	0.60a	0.54a	0.15a	0.47a	0.12a	0.05a	0.10a	0.03a	0.06a	0.02c	0.04a	0.02a	0.04a	0.01a	2.50a
Root	CK	32.87b	103.46c	81.73c	4.81a	71.24c	10.35c	4.92b	18.85c	2.31b	4.56b	2.46a	7.80b	0.93b	5.47b	0.79a	352.55c
	RS	46.70c	121.46c	90.58c	8.97b	84.75c	12.4c	5.48b	18.90c	2.74c	3.30b	7.91b	7.09b	1.05b	6.40c	0.90a	418.68c
	RSA	28.95a	83.80b	66.93b	1.50c	57.64b	6.22b	4.59a	12.24b	2.04b	1.61a	2.22a	7.90b	0.85b	5.07b	0.73a	282.29b
	BC	4.95a	21.93a	14.16a	1.54c	13.51a	1.75a	2.37c	5.75a	0.79a	0.02a	0.95b	2.70a	0.39a	2.41a	0.36b	73.58a

Note: The concentration unit of rare-earth elements in rice roots and shoots was $mg.kg^{-1}$, and the concentration unit in the grain of rice was $\mu g.kg^{-1}$. Data represent the average ($n = 3$); different lowercase letters on same line in the different parts of the rice indicate that the difference between treatment was significant ($p < 0.05$); RS refers to rice straw, RSA refers to rice straw ash (RSA), and BC refers to bone charcoal, CK refers to control check.

Table 3. Effects of Amendments on the Concentration of REEs (μg.L^{-1}) in Soil Solutions at Different Growth Stages in Rice

Period	Materials	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	∑REE
Tillering stage	CK	61.43c	51.54c	37.99c	9.74c	54.6bc	10.65c	2.35c	11.79c	1.19c	7.92c	1.53c	5.54c	0.69c	5.45c	0.80c	263.21c
	RS	63.73c	105.49d	63.65d	15.32d	78.60d	12.99c	2.72c	14.25c	1.38c	8.81c	1.64c	5.96c	0.69c	5.37c	0.78c	381.38d
	RSA	30.32b	24.44b	17.11b	4.29b	24.13b	4.71b	1.08b	5.33b	0.52b	3.36b	0.69b	2.61b	0.31b	2.48b	0.38b	121.76b
	BC	5.26a	3.57a	2.90a	0.83a	5.08a	1.01a	0.23a	1.01a	0.10a	0.67a	0.13a	0.50a	0.06a	0.48a	0.07a	21.90a
Heading period	CK	44.48c	35.41c	24.95c	6.41c	35.59c	6.76c	1.58d	7.41c	0.76c	4.90c	1.01c	3.75c	0.46c	3.58c	0.55c	177.60c
	RS	44.85c	57.00d	35.94d	8.79d	45.61d	8.54d	2.08c	8.90d	0.91d	5.80d	1.13c	4.07c	0.49c	3.75c	0.57c	228.43d
	RSA	23.43b	16.21b	12.30b	3.23b	18.55b	3.85b	0.92b	4.06b	0.40b	2.71b	0.56b	2.16b	0.27b	2.17b	0.33b	91.15b
	BC	2.59a	1.85a	1.76a	0.44a	2.60a	0.54a	0.12a	0.53a	0.05a	0.36a	0.07a	0.26a	0.03a	0.26a	0.04a	11.50a
Mature period	CK	15.80b	12.37c	8.59b	2.20b	12.23c	2.30b	0.54b	2.51b	0.26b	1.65b	0.35b	1.30b	0.16b	1.23b	0.19b	61.68b
	RS	23.13c	24.50d	16.02c	3.97d	20.77d	4.23c	1.12c	4.27c	0.45c	2.88c	0.57c	2.06c	0.25c	1.93c	0.30c	106.45c
	RSA	12.19b	7.76b	6.16b	1.65b	9.62b	2.05b	0.5b	2.1b	0.20b	1.43b	0.29b	1.15b	0.14b	1.19b	0.18b	46.67b
	BC	2.65a	1.12a	0.82a	0.22a	1.24a	0.38a	0.14a	0.33a	0.03a	0.27a	0.06a	0.24a	0.03a	0.33a	0.05a	7.91a

Note: Data is the average (n = 3), different lowercase letters in the same growing period of rice indicate that the difference between treatment is significant ($p < 0.05$); RS refers to rice straw, RSA refers to rice straw ash (RSA), and BC refers to bone charcoal, CK refers to control check.

Methods

Analysis of physical and chemical properties of soil samples

The soil pH was determined *via* the electrode method with a soil to water ratio of 2.5:1. The soil organic matter content was determined using the low-temperature external heat-potassium dichromate colorimetric method. The soil cation exchange concentration (CEC) was determined *via* the ammonium acetate method. The total concentrations of soil carbon, nitrogen, and sulfur were determined using an elemental analyzer (Vario EL III, Elementar, Frankfurt, Germany); and the texture composition was determined using a laser particle size analyzer (Zhuhai Omec Instrument Co., Ltd., Zhuhai, China).

Digestion of soil samples and determination of the total amount of REEs

In a quartz glass tube, 0.2 g soil samples were mixed with 5 mL aqua regia (HNO_3:HCl = 3:1). The mixture was incubated at room temperature overnight and then digested in an open digestion furnace the next day. The digestion temperature control program consisted of digestion at 90 ℃ or 30 min at 120 ℃ for 4 h, and at 140 ℃ until the soil turned white. After the digestion, the samples were placed in a ventilator to volatilize the acid and then transferred to 50-mL fixed-volume tubes. The samples were filled with ultrapure water to 50 mL, shaken well, filtered with a 0.45-μm filter membrane, and analyzed *via* inductively coupled plasma mass spectrometer (ICP-MS) (Agilent Technologies Inc., Palo Alto, CA, USA). The whole process of digestion was calibrated based on a national standard substance (GBW07405, National Standard Substance Research Center, Beijing, China).

Plant sample digestion and determination

A total of 0.2 g of the crushed plant sample was placed in a 50-mL polytetrafluoroethylene digestion tank, and then 3 mL high-grade pure nitric acid was added and mixed in evenly. The mixture was incubated at room temperature overnight, and then digested in a microwave digestion furnace (MARS5, CEM Microwave Technology Ltd., Matthews, NC, USA) the next day. First, the samples were heated to 120 ℃, which was maintained for 5 min, and then the temperature was increased to 160 ℃, which was maintained for 15 min. The whole process of digestion was controlled by a national standard substance (GBW08502, National Standard Substance Research Center, Beijing, China). After digestion, the samples were placed in a ventilated kitchen to cool and for acid removal, and then fixed to 40 mL, filtered through a 0.45-μm filter membrane, and finally analyzed *via* ICP-MS. The concentrations of P in the soil, rice roots, stems, and grain digestion solutions were determined *via* molybdenum-antimony resistance and ultraviolet-visible spectrophotometry (Shanghai Precision Science Instrument Co., Ltd., Shanghai, China).

Data analysis

The ability of rice roots to transfer rare earths to shoots is expressed by the translocation factor (TF) as follows: TF (%) = $C_{\text{Shoot-REE}}/C_{\text{Root-REE}} \times 100$.

The data were analyzed using Origin 9.0 (Origin Lab, Hampton, USA), SPSS 19.0 (SPSS Inc., Chicago, IL, USA), and Excel 2007 (Microsoft, Redmond, WA, USA) software, and the single factor variance (ANOVA) and Duncan test methods were used for significance analysis ($p < 0.05$). A bivariate correlation was used for the Pearson analysis.

RESULTS AND DISCUSSION

Effect of Different Modifiers on the Growth of Rice

According to Figs. 1 and 2, rice with the RSA and BC addition grew the best, and rice with the RS addition grew the worst. Rice straw, ash, and bone charcoal promoted rice growth in rare-earth-contaminated soil. The dry weights of the rice roots, shoots, and grains with RSA addition increased 124.5%, 91.3%, and 103.1%, respectively, compared with those of the CK treatment; and those with BC addition increased 101.8%, 82.6%, and 100.5%, respectively, compared with those of CK. The rice straw inhibited the growth of rice in the rare-earth-contaminated soil. The dry weights of the rice roots, shoots, and grains decreased 62%, 69.3%, and 82.5%, respectively, compared with those of CK.

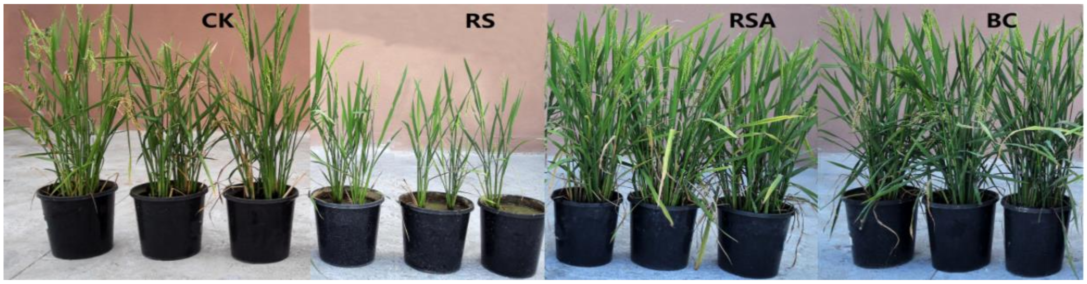

Fig. 1. The effect of different modifiers on the growth of rice (heading stage)

Effects of Different Modifiers on the Concentrations of Rare Earth Elements and P in Rice

Table 2 illustrates that the RS, RSA, and BC significantly affected the REE concentration in the roots of rice. The concentrations of REEs in the rice roots with the RS addition were significantly higher than those of the control; for instance, the concentrations of Ho, Pr, and Y were 221.5%, 86.5%, and 42.1% higher than those of CK, respectively. The average value of light REEs (La through Eu) increased 17.1%, and that of heavy REEs increased 24.9%. The total rare earth concentration of rice roots was reduced 19.9% compared to CK, whereas the concentrations of Pr, Dy, Sm, Y, La, Ce, Nd, Eu, and Gd as single REEs were significantly lower than those of the control ($P < 0.05$). In particular, the concentrations of Pr, Dy, and Sm were reduced 68.8%, 64.7%, and 39.9%, respectively. The effect of adding BC was the most significant.

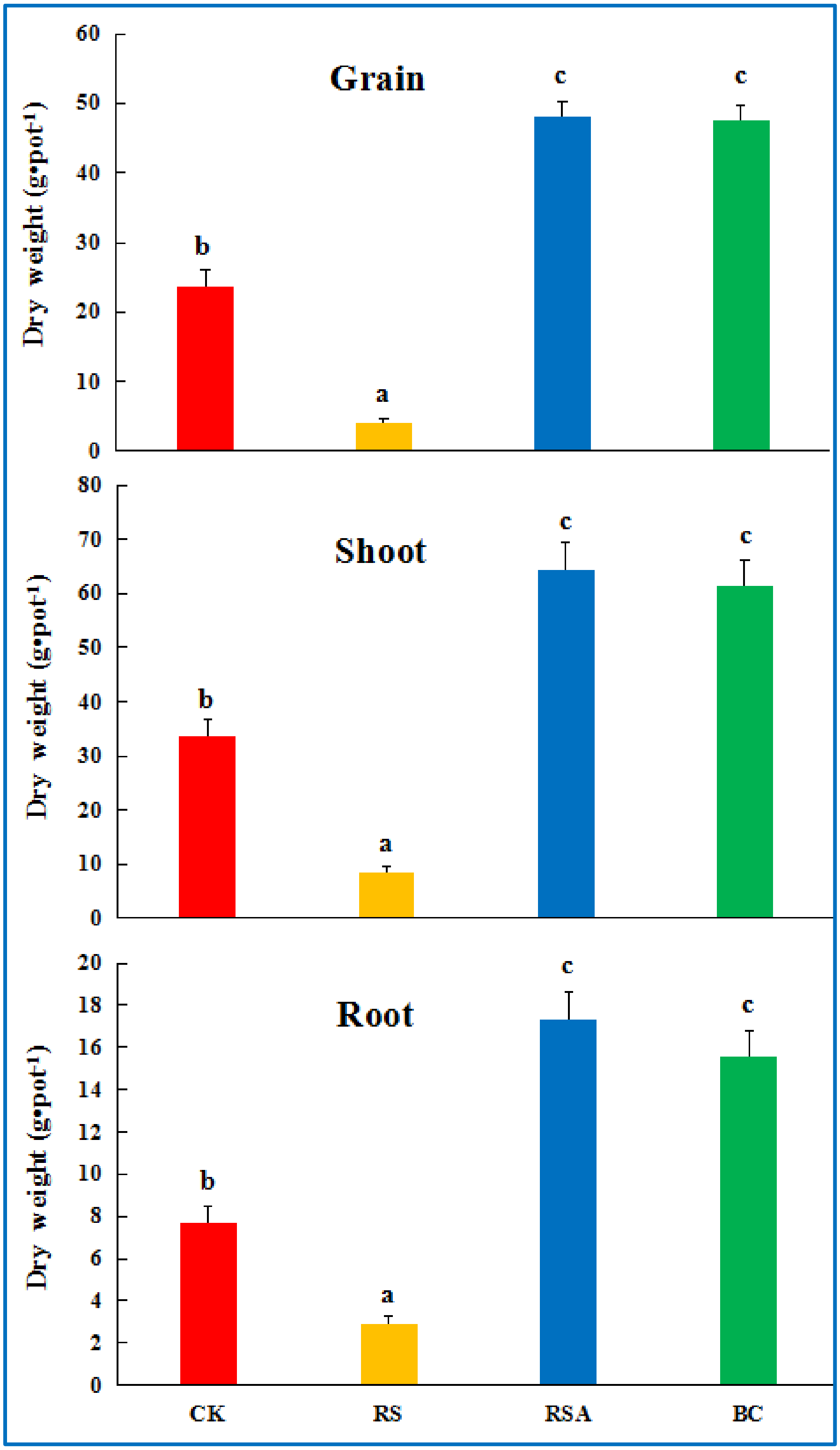

Fig. 2. Effects of different modifiers on biomass of rice; data represents the average (n = 3), and different lowercase letters in the same part of the same area indicate that the difference between treatments was significant ($p < 0.05$)

Table 4. Effect of Different Modifiers on Soil pH Value

	Planting Period	Rice Harvest
CK	5.06b	5.98a
RS	4.82a	6.06a
RSA	6.15c	6.08a
BC	6.82d	6.29b

Note: Data represent the average (n = 3), and different lowercase letters in the same growing period of rice indicate that the difference between treatments was significant ($p < 0.05$); RS refers to rice straw, RSA refers to rice straw ash (RSA), and BC refers to bone charcoal, CK refers to control check.

The concentration of all REEs in rice roots was significantly lower than that of the control ($P < 0.05$). The optimum elements were Dy, Y, and Sm, as they were 99.6%, 84.9%, and 83.1% lower, respectively, than those of the control. The average value of light REEs in rice roots decreased 80%, the average value of heavy REEs decreased 75.9%, and the total REE concentration was 79.1% lower than that of CK with the addition of BC.

The addition of RS, RSA, and BC also had a significant effect on the concentration of REEs in rice shoots (Table 2). The concentrations of most REEs in rice shoots were significantly lower than those in the control by adding RS, especially the concentrations of La, Ce, and Nd, which were 41.6%, 39.7%, and 37.5% lower than those of CK, respectively. Excluding Tm and Lu, the concentrations of the REEs in the leaves of rice with RSA addition were significantly lower than those of the control ($P < 0.05$). The largest reduction ratios were seen with Y, La, and Nd, which decreased 72.7%, 70.7%, and 70.7%, respectively, and the total REE concentration decreased 67.2%. When the BC was added, the concentrations in rice shoots of the 15 REEs studied were significantly lower than those of the control ($P < 0.05$), and the largest reduction ratios were found for La, Y, and Ce, which decreased 78.7%, 77.6%, and 77.3%, respectively. With BC addition, the average concentration of light REEs decreased 76.7%, the average concentration of heavy REEs decreased 73.1%, and the total REE concentration decreased 76%.

The addition of RS, RSA, and BC also affected the REE concentrations in grains. Adding RS significantly reduced the concentrations of Y, La, and Ce in grains, but it had no significant effect on the other REEs ($P > 0.05$). The RSA addition had no significant effect on the concentrations of the 15 REEs in the grains. With the addition of BC, the concentration of the15 REEs in rice grains was significantly lower than that of the control ($P < 0.05$). The largest reduction ratios were for Y, Sm, and La, which decreased 22.1%, 21.4%, and 20.8%, respectively. With BC addition, the average concentration of light REEs decreased 18.9%, the average concentration of heavy REEs decreased 17.3%, and the concentration of the total REEs decreased 18.3% (Table 2).

As shown in Fig. 3, the distribution of P concentration in rice plants was ordered as follows: roots > shoots > grains. The addition of RS, RSA, or BC increased the P concentrations of rice roots 123.4%, 249.7%, and 80.4%, respectively. When the BC was added, the concentrations of P in rice leaves and grains increased 215.5% and 29.6%, respectively, whereas the addition of RS or RSA had no significant effect on the P concentrations of rice stems and grains.

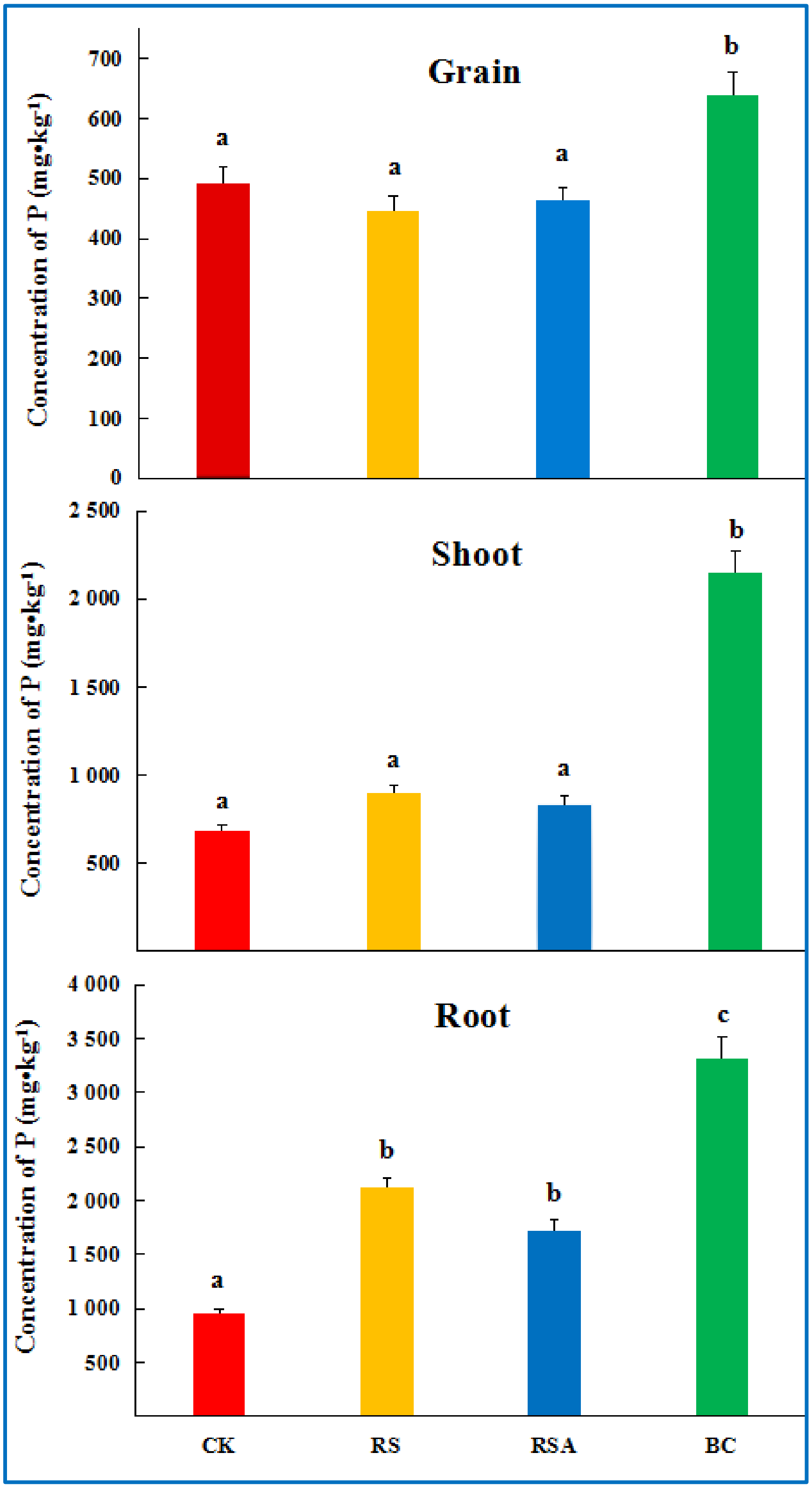

Fig. 3. Effects of different amendments on P concentration in roots, shoots, and grains of rice; data represents the average (n = 3); different lowercase letters in the same part of the rice indicate that the difference between treatment is significant ($p < 0.05$)

Effects of Different Amendments on the Concentration of Rare Earth Elements in Soil Solutions

Table 3 shows that the concentrations of La, Ce, Nd, and Pr in the soil solution with RS addition increased during the tillering stage, whereas the concentrations of the other REEs did not change significantly. The concentration of the 15 REEs in the soil solution with RSA addition was significantly lower than that of CK ($P < 0.05$). The largest reduction ratios were seen with Dy and Tb, which were 57.6% and 56.3% lower than those of CK, respectively. With RSA addition, the average concentration of light REEs decreased 54.6%, the average concentration of heavy REEs decreased 52.3%, and the total REE concentration decreased 53.7%. The effect of BC on reducing the concentration of REEs in the rice tillering period was very significant. The concentration of the 15 REEs in the soil solution was significantly lower than that of the control ($P < 0.05$). The largest reduction ratios were found in the La and Ce concentrations, which were 93.1% and 92.4% lower than those of CK, respectively, and the total REE concentration decreased 91.7%.

Table 5. Pearson Correlation Between REE Concentration and P Concentration in Rice Plant and REE Concentration in Soil Solution

	TSR	HSR	MSR	P Concentration in Grain	P Concentration in Shoots	P Concentration in Roots	PH1	PH2
REE Concentration in Grain	0.677	0.756	0.696	-0.93	-.999**	-0.937	-0.755	-.971*
REE Concentration in Shoots	.962*	.957*	0.877	-0.614	-0.638	-0.566	-.977*	-0.738
REE Concentration in Roots	0.937	.966*	0.948	-0.931	-0.905	-0.749	-0.947	-0.902

Note: TSR indicates rare earth concentration in soil solution during the tillering stage; HSR indicates rare earth concentration in soil solution at the heading stage, and MSR indicates rare earth concentration in soil solution at the mature stage; n = 5;
*Correlation is significant at the 0.05 level (2-tailed)
**Correlation is significant at the 0.01 level (2-tailed)

For rice in the heading stage, the concentrations of some of the soil solutions with added RS, such as La, Ce, and Pr, were significantly higher than those of CK, increasing 61%, 44%, 37.1%, and 29%, respectively, when compared with their concentrations with CK. The concentration of the 15 REEs with the RSA addition was significantly lower than that of CK ($P < 0.05$). The largest reduction ratios were found in the La, Ce, and Nd concentrations, which decreased 54.2%, 50.7%, and 49.6%, respectively. With RSA addition, the average concentration of light REEs decreased 50.3%, the average concentration of heavy REEs decreased 46.1%, and the concentration of total REEs decreased 48.7%. The addition of BC also caused the concentration of the 15 REEs to be significantly lower than that of the control ($P < 0.05$), for which the ratios of La and Y decreased 94.8% and 94.2%, respectively, and the concentration of total REEs decreased 93.5%.

For the rice in the maturity period, the concentration of the 15 REEs in the soil solution with added RS was significantly higher than that of CK ($P < 0.05$), and the highest ratios were seen with Eu, La, and Ce, which increased 107.4%, 98.1%, and 86.5%, respectively. With RS addition, the average concentration of light REEs increased 84.7%, the average concentration of heavy REEs increased 52.8%, and the concentration of total REEs increased 73%.

The concentrations of some REEs in the soil solution with RSA were significantly lower than those of CK, but the concentrations of other REEs did not change significantly. With the addition of RSA, the concentrations of La, Ce, and Nd greatly decreased, as they were 37.3%, 28.3%, and 25% lower than those of CK, respectively, and the total REE concentration decreased 24.4%. The concentration of the 15 REEs in the soil solution with BC was significantly lower than that of CK ($P < 0.05$). The largest reduction ratios were found in the La, Ce, and Nd concentrations, which were 91%, 90.5%, and 90% lower than those of CK, respectively. Moreover, with BC addition, the average concentration of light REEs was reduced 98.8%, the average concentration of heavy REEs by 83%, and the concentration of total REEs by 87.2%.

Effects of Modifiers on the Total REE Transport Coefficient (TF) in Rice

From Fig. 4, it can be seen that, compared with the CK, BC had no significant influence on the coefficient of the total rare earth transport of rice roots to the shoots ($P > 0.05$), whereas RSA and RS led to the total REE transport coefficients of rice being significantly lower than that of CK treatment, by 59% and 46.7%, respectively. This indicates that the RSA and RS treatments significantly inhibited the transfer of total dilute soil elements from the underground part of rice to the shoots.

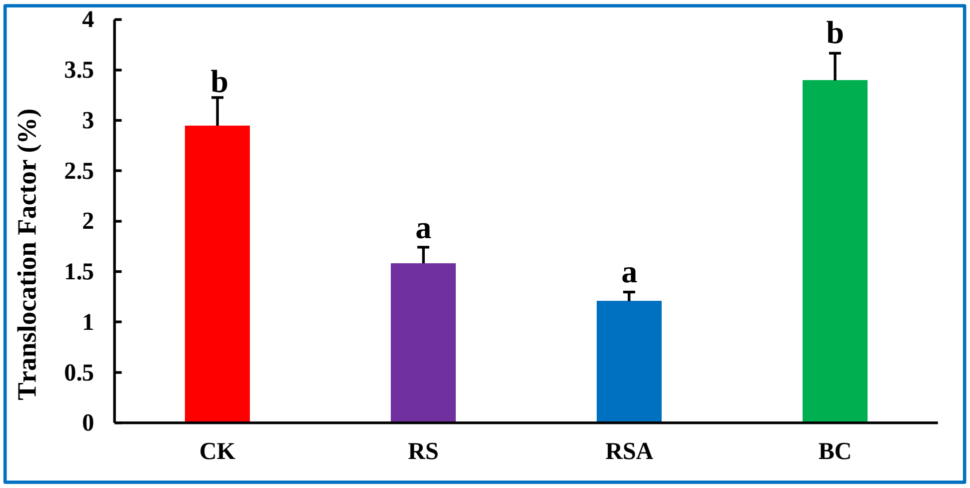

Fig. 4. Effects of different amendments on total REE transport in rice; data represent the average ($n = 3$), and different lowercase letters indicate that the difference between treatment is significant ($p < 0.05$)

The addition of RS, RSA, or BC significantly affected the concentrations of REEs in the rice roots, shoots, and seeds. The concentrations of REEs in rice roots, shoots, and grains were significantly positively correlated with the concentrations of REEs in the soil solution at the tillering, heading, and maturity stages, and showed significantly negative correlations with the concentrations of P in rice roots, shoots, and grains and with soil pH

(Table 5). Due to the difference in the number of electrons in the 4f orbitals of different REEs and the difference between atomic radius and ionic radius, the effects of different elements on the light and heavy REEs in the soil solution differed, which affected the growth of rice differently. The addition of RS led to less rice growth than with CK, probably because of the decrease in soil pH (Table 4) during the period of RS addition. This resulted in a significant increase in the concentration of REEs in the soil solution, thus causing the rice roots to absorb more rare earths, which affected the absorption and transport of rice to P and inhibited the growth of rice. After adding RSA or BC to the soil, the pH of the soil increased and the concentration of REEs in the soil solution was significantly reduced. This promoted the rice capacity of absorption and transport of phosphorus (P), thus reducing the concentration of REEs in the roots, stems, and grains of rice, promoting the growth of rice, and increasing the yield of rice.

Returning rice straw to the fields will change the physical and chemical properties of the soil and affect the bio-availability of REEs in the soil. In this study, it was found that adding rice straw to soil caused the pH to decrease but the DOM concentration to increase, and the concentration of soil-soluble REEs increased significantly at the rice tillering, heading, and maturity stages. Some studies have shown that adding rice straw can significantly reduce the Cd concentration in grains. This is because rice straw is rich in silicon and sulfur (Konboon *et al*. 2000; Seyfferth *et al*. 2013). Returning rice straw to the fields can increase the concentrations of available silicon and available sulfur in soil. Silicon can promote the fixation of Cd in plant cell walls (Liu *et al*. 2013), and sulfur can promote the isolation of Cd from plant vacuoles. By increasing the immobilization of cell walls or vacuolation, the transport of Cd to the upper part of rice is reduced and the accumulation of Cd in the grain is also reduced. In the current study, the addition of RS resulted in a reduction in the coefficients of transport of rare earths from rice roots to shoots and to grains and from rice leaves to grains, which were reduced 5.6%, 18.6%, and 13.9%, respectively, compared with CK. This may have been due to the increases of Si and S concentrations in the rice roots and shoots after RS addition and increase in the fixation of REEs in the cell walls of rice. Promoting the segregation of REEs by vacuoles in rice resulted in a significant decrease in the transport of REEs into grains.

After adding RSA, the concentrations of REEs in the soil solution of the rice tillering, heading, and maturity stages were significantly lower than those of CK, and the concentrations of REEs in the roots, stems, and grains of rice were significantly reduced. Moreover, the concentrations of P in the roots and stems of rice were significantly higher than those of CK. The addition of RSA significantly reduced the coefficient of REEs that were transferred from rice roots to shoots, which promoted the growth of rice and increased the biomass and yield of rice significantly. The concentration of silicon dioxide in the RSA was higher than 70%, which is rich in essential nutrients such as K and P (Ma *et al*. 2002). The addition of RSA increased the pH of the soil (Table 4).

Many studies have shown that siliceous ions react with heavy metal ions to form silicic acid precipitates after the silicon-containing materials are added to soil. The pH of soil can be increased, the ability to adsorb heavy metal ions and the activity of heavy metals in soil can be enhanced, and the activity of heavy metals in soil is reduced (Ma and Takahashi 2002). Silicon is an important element of cell walls. Silicon and REEs can be attached to the cell wall of plant cells. Therefore, the addition of silicon can strengthen the intercepting effect on the REEs of the plant cell wall, thus reducing the transfer of REEs to other plant parts (Ding *et al*. 2007).

The chemical formula of bone charcoal is $Ca_3(PO_4)_2$, and Ca_3CO_3 and $Ca_3(PO_4)_2$ accounted for 80%. Bone charcoal is alkaline, with alkali ions and a large specific surface area (Rinklebe *et al*. 2016; Martins *et al*. 2017). The concentrations of REEs in the roots, shoots, and grains of rice were significantly lower than those of CK (Table 2). At the same time, the concentrations of REEs in the soil solutions at the tillering, heading, and maturity stages of rice were also significantly lower than those of CK (Table 3). The addition of bone charcoal promoted the absorption and accumulation of P fertilizer, promoted the growth of rice, and increased the biomass and yield of rice. The concentrations of REEs in the rice roots, shoots, and grains had a significant negative correlation with the soil pH and plant P concentration but had a significant positive correlation with the concentration of REEs in the soil solution (Table 5). The reasons for the reduction in the concentration of REEs in the soil solution and the promotion of growth and development of rice are as follows. (1) By adding bone charcoal to increase the pH of soil, the electrostatic adsorption reaction of the REEs and clay particles is inhibited, and some metal hydroxides are easily generated (Chen 2012). (2) Bone charcoal can be associated with pH. The surface charge density of the soil particles affects the affinity of the soil to the REEs. With an increase in the pH of the solution, the desorption of the REEs in the soil gradually decreases (Cao *et al*. 2001; Rees *et al*. 2014). (3) The bone charcoal added to the soil not only can directly adsorb some REEs, but it also can increase the negative charge on the surface of the soil particles, which can enhance the adsorption of the cations (Naidu *et al*. 1994; Chen *et al*. 2010), thus reducing the activity and mobility of the soil REEs. (4) The increase in the pH caused by the addition of bone charcoal can change the morphological distribution of organic ligands in soil, thus influencing the desorption of REEs (Peng An *et al*. 2003). (5) The metal ions in the soil can form a variety of complexes with the organic and inorganic coordination bodies. With an increase in the pH of the soil, the negative power of the COO-, OH-, and C=0 groups on the surface of the organic matter increases, and metals are separated, and their complexation ability is also enhanced, thus leading to the formation of multiple complexes (chelates) (Chen *et al*. 2003; Mignardi *et al*. 2012; Zhao and Wilkinson, 2015). As a result, the concentration of REEs in the soil solution is reduced. (6) After the bone charcoal is added to the soil, the phosphate ions produced *via* the soil acid can form a refractory metal phosphate precipitate with the REE ions, which may be the main reason for the reduction of the REE concentration in the soil solution *via* BC addition (Cao *et al*. 2009; Buda *et al*. 2010; Wang and Liang 2014).

CONCLUSIONS

1. Of the three different modifiers, the addition of straw was not conducive to the growth of rice in the rare-earth-contaminated soil. The addition of 2.5% straw resulted in a reduction of the rice roots, shoots, and dry weight of grains of 62%, 69.3%, and 82.5%, respectively, compared with those of CK. Adding RSA and BC can promote the growth and development of rice. When adding 2.5% RSA to the rare-earth-contaminated soil, the rice roots, shoots, and the dry weight of grains increased 124.5%, 91.3%, and 103.1%, respectively, compared with those of CK. Adding 2.5% BC resulted in the reduction of rice roots, shoots, and the dry weight of grains by 101.8%, 82.6%, and 100.5%, respectively, compared with those of CK.

2. The concentrations of total REEs in the rice roots, stems, and grains were lower than those of CK with the BC addition, and the concentrations of total REEs in the rice roots and shoots were significantly reduced with the RSA addition. However, the influence on the total REE concentration in grains was not obvious. Adding RS could significantly reduce the total REE concentration in rice stems and roots, but had no significant effect on the concentration of total REEs in the grains and rice roots. The concentrations of the 15 REEs in the rice roots, stems, and grains were decreased significantly with the addition of BC when compared with the control group. However, the addition of RS or RSA significantly reduced the concentration of some REEs in rice plants, but it had no significant effect on the concentration of some REEs.
3. The addition of RS caused the total REE concentration of the soil solutions at the tillering, heading, and maturity stages of rice to be significantly higher than those of CK, whereas adding RSA or BC caused them to be significantly lower than those of CK. The concentrations of the 15 REEs in the soil solutions at the tillering, heading, and maturity stages of rice were significantly lower than those of the control group with BC addition, whereas BC treatment significantly improved the absorption of P by rice.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (41561096; 41867062), and the Jiangxi Science and Technology Project of China (20142BAB203026).

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Article submitted: July 19, 2018; Peer review completed: September 22, 2018; Revised version received and accepted: September 25, 2018; Published: October 2, 2018.
DOI: 10.15376/biores.13.4.8593-8613