Environmental Science and Pollution Research (2020) 27:40001–40008
https://doi.org/10.1007/s11356-020-10028-3

RESEARCH ARTICLE

Effects of steel slag amendments on accumulation of cadmium and arsenic by rice (*Oryza sativa*) in a historically contaminated paddy field

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Received: 1 May 2020 / Accepted: 6 July 2020 / Published online: 10 July 2020

Abstract

Paddy soil contamination by cadmium (Cd) and arsenic (As) is a great concern. Field experiments were conducted to study the effects of steel slag (SS, 2.0 and 4.0 t ha^{-1}) on the solubility of Cd and As in soil and their accumulation by rice plants grown in a historically co-contaminated paddy field with Cd and As. The results showed that SS amendment (4.0 t ha^{-1}) significantly decreased soluble concentrations of Cd in pore-water but increased that of As, related to markedly elevated soil pH and soluble silicon, phosphorus of pore-water in rice rhizosphere at both heading and mature stages. The amendments also evidently decreased Cd but enhanced As in iron plaque on root surfaces, while the formation of iron plaque was not significantly increased. Further, SS amendment (4.0 t ha^{-1}) markedly reduced Cd concentrations in rice tissues (roots, straw, and brown rice) by 48–78% at both stages, though increased As by 13–38%. Cadmium translocation from roots to aerial parts decreased significantly after the amendments, but not for As. Besides, SS application increased the biomass of roots, straw and grains, and root antioxidant enzyme activities. Collectively, steel slag decreased Cd accumulation in rice tissues and in iron plaque but increased those of As, likely due to steel slag decreasing soluble Cd and enhancing soluble As in pore-water, related to soil pH and soluble nutrients (Si, P), and restraining Cd translocation within rice. Our results indicate that steel slag represents a favorable potential for Cd-contaminated paddy soils, though it seems undesirable for Cd and As co-contamination.

Keywords Steel slag · Paddy rice · Pore-water · Iron plaque · Cadmium attenuation · Arsenic enhancement

Introduction

During the past few decades, soil contamination by heavy metals has become a serious issue in agriculture, mainly due to anthropogenic activities, such as mining, smelting operations, sewage irrigation, and fertilizer application (Rai et al. 2019; Raj and Maiti 2020). Regarding heavy metals, particularly cadmium (Cd) and arsenic (As, a metalloid but usually recognized as a heavy metal) are extremely hazardous pollutants in agricultural soils, giving rise to suppression of crop growth, excessive Cd or As in cereals, and threats to human health through food consumption (Rai et al. 2019; Raj and Maiti 2020). Rice (*Oryza sativa* L.) is one of the foremost cereal crops over the globe, but has a high ability to uptake Cd and As from soils and thus has become a major dietary source of Cd and As (Irshad et al. 2020). Geochemical behaviors of Cd and As vary greatly under paddy field condition, generating a major risk for rice cultivation (Irshad et al. 2020). To ensure safe cereal production, the remediation of paddy soils contaminated by Cd and As is imminently needed. One widely employed technology is in situ chemical stabilization of heavy metals by adding soil amendments (Kumpiene et al. 2019).

Steel slags, consisting of main constituents of silicon (Si), calcium (Ca), phosphorus (P), iron (Fe), and manganese (Mn), are low-cost, abundant, and alkaline byproducts from steel mills (Navarro et al. 2010; Guo et al. 2018). In some countries, steel slag has been maturely used as silicate fertilizer, phosphate fertilizer, and soil modification in agriculture (Guo et al.

Responsible Editor: Elena Maestri

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2018). Steel slag application improves soil quality (León-Romero et al. 2018), increases soil nutrient availability and rice yield (Wang et al. 2015a; He et al. 2017; Wang et al. 2018a), and reduces methane emission in paddy fields (Wang et al. 2015a; Wang et al. 2018b). Further, steel slag amendment can immobilize heavy metals (e.g., Cd and lead) in soil and decrease their concentrations in crops (Kim et al. 2012; Ning et al. 2016; He et al. 2016; Hu et al. 2019), but the amendment may have positive or negative impacts on As uptake by plants (Nejad et al. 2017; León-Romero et al. 2018). Nevertheless, information is lacking regarding steel slag for the remediation of co-contaminated paddy soils with Cd and As, and the possible mechanisms behind steel slag effects on the accumulation of Cd and As in crops are poorly understood.

Heavy metals uptake by plants depends mainly on the solubility and availability of these metals in soil (Zeng et al. 2011). The heavy metals measured in soil pore-water denote the most soluble fractions in soil for plant uptake (Beesley et al. 2010; Moreno-Jiménez et al. 2011; Concas et al. 2015) and are good indicators to assess the remediation efficiency of soil amendments (Zheng et al. 2012; Beesley et al. 2014). Schemes of soil amendments may influence the solubility of Cd and As in soil by the modification of chemical properties altering soil pH and binding forms of metals in soil solution (Beesley et al. 2014). Solubility of heavy metals is also affected by the application of beneficial nutrients such as Si and P, owing to their impacts on adsorption of metals onto soil and metal bioavailability for crop uptake (Sarwar et al. 2010). It is possible that alkaline steel slag containing Si- and P-rich compounds can change the aforementioned soil factors, therefore affecting the accumulation of Cd and As in rice. In addition, iron plaque is generally formed on rice root surfaces and can modify the uptake and accumulation of metal(loid)s in rice plants (Liu et al. 2004; Wang et al. 2013; Cheng et al. 2014). The formation of iron plaque and its adsorption capacity on metals are affected by soil conditions and soil amendments (Zheng et al. 2012). As reported, the biochar amendments increased iron plaque formation on the surfaces of rice roots and sequestrated more Cd and As in iron plaque (Irshad et al. 2020), whereas the Cd concentrations in iron plaque decreased (Zheng et al. 2015). It is hypothesized that supplementing paddy soils with steel slag may affect iron plaque formation and thereby the uptake of Cd and As by rice; however, this potential effects are unclear.

Based on the above observations and hypothesis, this work aimed to study the effects of steel slag on the solubility of Cd and As in soil and their accumulation and translocation within rice plants grown in a historically co-contaminated paddy field with Cd and As, and to explore the possible mechanisms involved. Paddy field (rhizobag) experiments were carried out to explore the effects by determining soil pH; soluble Cd, As, Si, and P in soil pore-water; concentrations of Cd, As, and Fe in iron plaque; concentrations of Cd and As in rice tissues (roots, straw, and brown rice); and translocation of Cd and As from roots to aerial parts. Rice biomass and root antioxidant enzyme activities were also measured.

Materials and methods

Experimental location, steel slag, and rice variety

The experimental site was established at a paddy field in Shangba village (24°27′N; 113°48′E) close to Dabaoshan mine, Guangdong Province, southern China. The area has a subtropical humid climate with an average annual rainfall of 1762 mm and an average annual temperature of 20.3 °C. Dabaoshan mine is a cluster of opencast multi-metal mines initiating operation in the 1960s and is still in operation. The studied paddy field has been chronically contaminated due to the acid mine drainage from Dabaoshan mine. The total concentrations of Cd and As in the paddy soil (Table 1) were, respectively, 8.2-fold and 2.6-fold above the Soil Environmental Quality – Risk Control Standards for Soil Contamination of Agricultural Land (Cd 0.3 mg kg^{-1} and As 30 mg kg^{-1}, in GB 15618–2018 of China). Thus, the soil markedly exceeded the national risk control levels for Cd and As in soil and was considered relatively highly contaminated. In this study, steel slag powders through a 0.15-mm sieve were obtained from Shaoguan Steel Group Company of Guangdong Province (He et al. 2016). The slag was strongly alkaline and has very low concentrations of Cd and As (Table 1), and could be considered safe for use in agricultural soils referring to the maximum allowable content of Cd and As in the Control Standards of Pollutants in Fly Ash for Agricultural Use (Cd 5 mg kg^{-1} and As 75 mg kg^{-1}, in GB 8173–87 of China). Seeds of rice cultivar (*Oryza sativa* cv.

Table 1 Properties of the paddy soil (mean ± SE, $n = 5$) and steel slag (mean ± SE, $n = 4$)

Particulars	Paddy soil	Steel slag
pH (1:2.5 w/v water)	4.7 ± 0.04	11.8 ± 0.14
Total Cd (mg kg^{-1})	2.46 ± 0.07	0.06 ± 0.002
Total As (mg kg^{-1})	79 ± 4.2	1.3 ± 0.02
Organic C (g kg^{-1})	17 ± 0.6	
Total P (g kg^{-1})	0.55 ± 0.02	6.8 ± 0.10
Total K (g kg^{-1})	0.71 ± 0.03	0.09 ± 0.003
Total Fe (g kg^{-1})	36 ± 0.8	82 ± 1.0
Total Mn (g kg^{-1})	0.17 ± 0.02	1.9 ± 0.04
Total Ca (g kg^{-1})	1.25 ± 0.04	216 ± 9.0
[a]Extractable Si (mg kg^{-1})	48 ± 1.7	945 ± 42

[a] Extractable Si extracted using Mehlich 3

Tianyou 122) were from the Rice Research Institute of Guangdong Academy of Agricultural Sciences and were used in field experiments.

Paddy field (rhizobag) experiments

A total of 12 experimental plots were set up with each plot having an area of 6 m^2 (2 m × 3 m), 30-cm ridges around each plot, and 20-cm spacing between adjacent plots. The plots were randomly divided into three treatments, each with four replicates. The treatments included control (without steel slag) and steel slag amendments with the addition (low and high dosages) of 2.0 t slag ha^{-1} (SS1) and 4.0 t slag ha^{-1} (SS2), according to our previous study (He et al. 2016). Steel slag was added into each plot and mixed thoroughly with the soils of a 15-cm tillage layer. Subsequently, about 1.0 kg of the mixed soils in each plot were added to a cylindrical rhizobag (made of 30-μm nylon mesh, 12-cm diam, 15-cm height) that was designed to differentiate rhizosphere from non-rhizosphere zones (Cheng et al. 2014; Wang et al. 2015b). The interface between soils and plant roots is the rhizosphere that is the most active site for direct material exchange in soil-plant systems (Wang et al. 2015b). Thus, the effects of steel slag on paddy soils, rice plants, and their possible relationships may be availably explored using rhizosphere bag. Four rhizobags were placed into the central zone of each plot, prior to transplanting 20-day-old seedlings which were first cultivated in a seedbed at an unpolluted field. Two rice seedlings were transplanted to a rhizobag that was gently taken from the field at harvested stages, and the transplantation of the rest rice seedlings followed the conventional way in each plot. Rice plants were cultivated for 90 days and were harvested at heading and mature stages. All experimented plots followed the same agricultural managements, such as irrigation with clean water, fertilization, and weeding (He et al. 2016).

Sampling and analytical methods

Samples of rice plants harvested were separated into roots and straw at the heading stage, and roots, straw, and grains at the mature stage. Straw and grains were oven-dried at 50 °C until a constant weight for biomass. Roots were taken from rhizobag soils by gentle sieving for further measurements. The activities of antioxidant enzymes (catalase (CAT), superoxide dismutase (SOD), and ascorbate peroxidase (APX)) of fresh roots gathered at the heading stage were determined as reported by Liu et al. (2013). Iron plaque on root surfaces was extracted using dithionite-citrate-bicarbonate (DCB) solution (Otte et al. 1989; Liu et al. 2004). After washing with deionized water, rice roots were incubated in 40 ml of DCB solution for 60 min, and then were rinsed three times with deionized water. Rinsed water was transferred to the DCB extracts and the final solution was made up to 50 ml for analysis of Cd, As, and Fe in iron plaque. Subsequently, the dried samples of roots, straw, and grains were pretreated and analyzed for Cd and As according to the methods of Wang et al. (2013).

Soil samples were collected from the rhizobags and were air-dried. Soil pH was measured in a slurry with a water–solid ratio of 2.5:1 using a pH meter (pH 510, Eutech Instruments, Singapore). Soil pore-water was collected by in situ pore-water samplers (Rhizosphere Research Products, the Netherlands) inserted into the base of the rhizobags, referring to the method described by Kidd et al. (2007) and Zheng et al. (2012). The pore-water was sampled at both heading and mature stages and concentrations of Cd, As, Si, and P in pore-water were determined. Elemental concentrations in samples were measured using graphite furnace atomic absorption spectrophotometer (GFAAS, Hitachi Z-2000, Japan) for Cd, atomic fluorescence spectrometry (AFS, Beijing, Jitian Instrument Co., Ltd.) for As, and inductively coupled plasma optical emission spectrometry (ICP-OES, Optima 2000 DV, Perkin Elmer, USA) for Fe, Si, and P. Blanks, soil standard material (GBW-07435), and rice standard material (GBW-10045) (China Standard Materials Research Center, Beijing, China) were performed for quality control.

Statistical analysis

Translocation factors for Cd or As from roots to aerial parts were calculated as follows: TF = Cd or As concentrations in aerial parts/roots. Statistical analyses were performed using SPSS (version 18.0) software for Windows. Results were presented as arithmetic means with standard error attached, and means were compared using one-way variance (ANOVA) followed by the least significant difference (LSD) test at 5% probability level.

Results

Steel slag effects on soil pH and pore-water Cd, As, Si, and P

Steel slag amendments of 2.0 t ha^{-1} (SS1) and 4.0 t ha^{-1} (SS2) showed beneficial effects on pH and soluble Cd (but not As), Si, and P in rhizosphere soils at both the heading and mature stages of rice, and the effects were greater in SS2 treatment than those in SS1 treatment (Fig. 1). Compared with the unamended controls, both treatments decreased soluble Cd concentrations in pore-water, and especially SS2 treatment resulted in significant ($P < 0.05$) reductions by 45% and 34% at the heading and mature stages, respectively (Fig. 1). In contrast to the trend for Cd, both treatments enhanced As concentrations in pore-water, with SS2 affecting significant ($P < 0.05$) increases by up to 2.5-fold at both stages (Fig. 1).

In addition, both SS1 and SS2 treatments promoted significant increases in soil pH in excess of 1.1–2.5 units ($P < 0.05$), and also

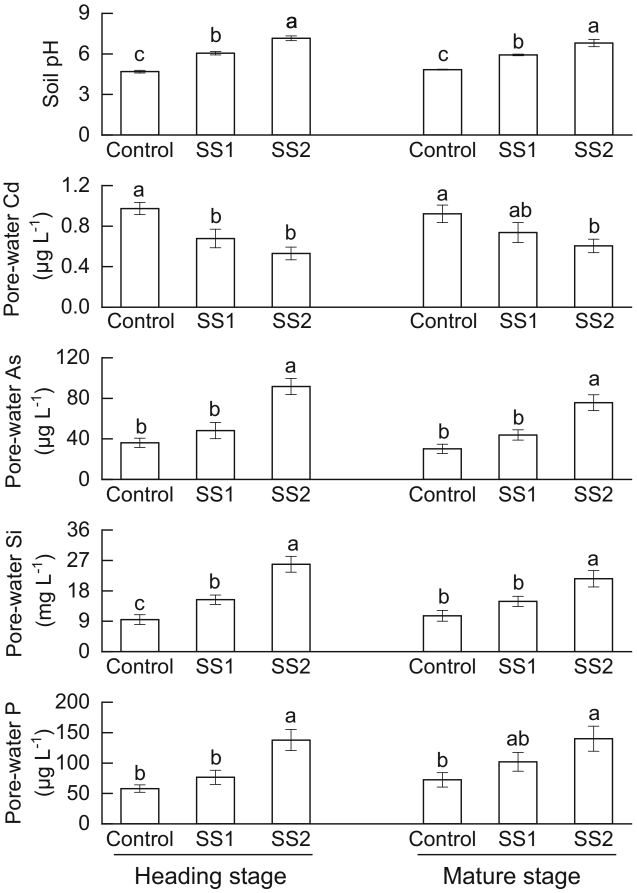

Fig. 1 Rhizosphere soil pH and pore-water concentrations of Cd, As, Si, and P in paddy field experiments with and without steel slag (mean ± SE, $n = 4$). SS1: steel slag (2.0 t ha^{-1}), SS2: steel slag (4.0 t ha^{-1}). Different letters within the same parameter and the same stage indicate significant difference ($P < 0.05$) between the treatments

improved soluble concentrations of Si and P in pore-water at both stages, particularly for SS2 causing significant ($P < 0.05$) increases in pore-water Si and P by 1.9–2.7-fold (Fig. 1).

Steel slag effects on Cd, As, and Fe in iron plaque

Compared with the controls, the formation of iron plaque (DCB-Fe) on rice root surfaces was not significantly increased by steel slag amendments at both the heading and mature stages (Fig. 2). The influence of steel slag varied on concentrations of Cd and As in iron plaque. Both treatments significantly ($P < 0.05$) decreased Cd concentrations in iron plaque (DCB-Cd) at both stages, while the concentrations of As in iron plaque (DCB-As) increased markedly ($P < 0.05$) at the heading stage (Fig. 2).

Steel slag effects on Cd and As accumulation and translocation in rice

As expected, the Cd concentrations in rice tissues (roots, straw, and brown rice) decreased after steel slag amendments at the heading and mature stages, and the reductions were more in SS2 treatment than those in SS1 treatment (Table 2). Compared with the controls, SS2 treatment significantly ($P < 0.05$) reduced the concentrations of Cd in rice tissues by 48–78% at both stages, and the concentrations of Cd in brown rice were lower than 0.2 mg kg^{-1} (the limit of the Food Quality Standard GB2762–2017 of China). Furthermore, both treatments decreased the translocation factors of Cd from roots to aerial parts at both stages, and significant differences ($P < 0.05$) were observed at the heading stage (Table 2).

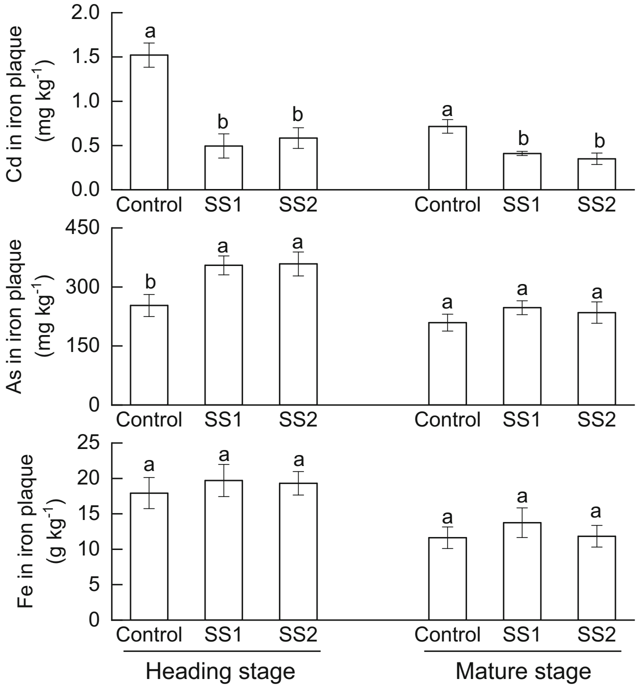

Fig. 2 Concentrations of Cd, As, and Fe in iron plaque on rice root surfaces in paddy field experiments with and without steel slag (mean ± SE, $n = 4$). SS1: steel slag (2.0 t ha^{-1}), SS2: steel slag (4.0 t ha^{-1}). Different letters within the same parameter and the same stage indicate significant difference ($P < 0.05$) between the treatments

In contrast to the Cd concentrations, steel slag amendments increased the concentrations of As in rice tissues (roots, straw, and brown rice) by 13–38% at the heading and mature stages, and notably, SS2 treatment resulted in significant ($P < 0.05$) increments (29–38%) in roots at both stages and in straw at the heading stage, compared with the controls (Table 2). No significant change in As translocation from roots to aerial parts existed after steel slag amendments (Table 2).

Steel slag effects on rice biomass and root antioxidant enzyme activity

Compared with the controls, steel slag amendments enhanced the dry biomass of rice tissues (roots, straw, and grains) at both the heading and mature stages, and significant ($P < 0.05$) increases in plant biomass (roots, straw, and grains) by SS2 treatment ranged from 24 to 40% (Table 3). Besides, steel slag

Table 2 Concentrations (conc., mg kg^{-1}) of Cd and As in rice tissues (roots, straw and brown rice) and their translocation factors from roots to aerial parts in paddy field experiments with and without steel slag (mean ± SE, $n = 4$)

Metal	Treatment	Heading stage			Mature stage			
		Conc. in roots	Conc. in straw	Translocation factor	Conc. in roots	Conc. in straw	Conc. in brown rice	Translocation factor
Cd	Control	5.6 ± 0.7a	0.88 ± 0.12a	0.16 ± 0.005a	7.3 ± 1.1a	1.82 ± 0.14a	0.31 ± 0.03a	0.30 ± 0.03a
	SS1	3.1 ± 0.4b	0.37 ± 0.04b	0.12 ± 0.004b	4.9 ± 0.4b	1.15 ± 0.14b	0.19 ± 0.02b	0.27 ± 0.01a
	SS2	1.6 ± 0.2c	0.19 ± 0.03b	0.12 ± 0.005b	2.9 ± 0.3b	0.61 ± 0.10c	0.16 ± 0.01b	0.26 ± 0.03a
As	Control	157 ± 16b	9.3 ± 0.9b	0.059 ± 0.002a	142 ± 8.0b	11.8 ± 1.2a	0.16 ± 0.01a	0.084 ± 0.005a
	SS1	177 ± 15ab	11.1 ± 1.3ab	0.061 ± 0.005a	159 ± 16ab	13.3 ± 1.6a	0.18 ± 0.01a	0.084 ± 0.005a
	SS2	215 ± 18a	12.7 ± 1.0a	0.059 ± 0.001a	183 ± 9.0a	14.7 ± 1.1a	0.18 ± 0.02a	0.082 ± 0.003a

SS1 steel slag (2.0 t ha^{-1}), *SS2* steel slag (4.0 t ha^{-1}). Different letters within the same parameter and the same stage indicate significant difference ($P < 0.05$) between the treatments

amendments slightly increased the activities of the antioxidant enzyme SOD in rice roots, but the activities of CAT and APX increased significantly ($P < 0.05$) by SS2 treatment (Table 3).

Correlations between the parameters

The linear regression analysis revealed that, after steel slag amendments, the concentration of Cd in pore-water was negatively correlated with soil pH ($R = -0.993$; $P < 0.01$), pore-water Si concentrations ($R = -0.953$; $P < 0.01$), and pore-water P concentrations ($R = -0.876$; $P < 0.05$), but pore-water As was positively correlated with soil pH ($R = 0.932$; $P < 0.01$), pore-water Si ($R = 0.983$; $P < 0.01$), and pore-water P ($R = 0.911$; $P < 0.05$) (Table 4). Significantly positive correlations were also found between pore-water Cd and Cd concentrations in roots ($R = 0.918$; $P < 0.01$) and between pore-water As and As concentrations in roots ($R = 0.950$; $P < 0.01$) (Table 4).

Discussion

Excessive accumulation of Cd and As in crops has become a pressing concern. The present study demonstrated that steel slag amendment significantly increased rice production and decreased Cd concentrations in rice tissues and in iron plaque, though enhanced rice concentrations of As (Table 2), being a new suggestion that steel slag seemed impossible to simultaneously mitigate the accumulation of Cd and As in rice plants. To our knowledge, it is the first to investigate the effects of steel slag on simultaneous alteration of Cd and As in iron plaque and their accumulation in rice plants grown in a historically co-contaminated paddy field with Cd and As. Most previous studies in steel slag application have been limited to glasshouse or laboratory conditions, and the results may therefore not directly predict the field behavior. Thus, a discrimination of mechanisms by steel slag controlling the mobility of Cd and As from soils to rice plants enables the benefit of steel slag application under field environment.

The contrasting effects of steel slag on As and Cd in rice plants may be related to the amendment impact on soil geochemical properties. Steel slag amendment clearly decreased soluble Cd concentrations of pore-water in rhizosphere soils but increased that of As, compared with the controls (Fig. 1). The changes of Cd and As in pore-water are in consistence with the alkaline biochar study by Zheng et al. (2012) and Beesley et al. (2014). Possibilities below can be envisaged to explain the decrease of Cd solubility in soil by steel slag amendments. It is generally recognized that soil pH is a key factor regulating Cd concentrations in soil solution, and an elevated soil pH can reduce soluble Cd by increasing

Table 3 Biomass of rice tissues (roots, straw and grains) (kg plot^{-1} dry weight) and activities of antioxidant enzymes, catalase (CAT, u g^{-1} min), ascorbate peroxidase (APX, u g^{-1} min), and superoxide dismutase (SOD, u g^{-1}) of fresh rice roots in paddy field experiments with and without steel slag (mean ± SE, $n = 4$)

s	Heading stage					Mature stage		
	Roots	Straw	CAT	APX	SOD	Roots	Straw	Grains
Control	0.31b ± 0.03b	2.53 ± 0.14b	49 ± 5.3b	119 ± 12b	84 ± 8.4a	0.22 ± 0.02b	1.83 ± 0.12b	3.06 ± 0.16b
SS1	0.39 ± 0.02ab	3.37 ± 0.06a	59 ± 7.4ab	188 ± 12a	92 ± 8.0a	0.25 ± 0.02ab	2.49 ± 0.14a	3.63 ± 0.23ab
SS2	0.41 ± 0.03a	3.44 ± 0.17a	73 ± 3.9a	187 ± 9.1a	92 ± 14a	0.30 ± 0.04a	2.57 ± 0.18a	3.79 ± 0.24a

SS1 steel slag (2.0 t ha^{-1}), *SS2* steel slag (4.0 t ha^{-1}). Different letters within the same parameter and the same stage indicate significant difference ($P < 0.05$) between the treatments

Table 4 Correlation coefficients (R) between concentrations (μg L^{-1}) of Cd or As in pore-water and soil pH, pore-water Si concentrations (mg L^{-1}), pore-water P concentrations (μg L^{-1}) and concentrations (mg kg^{-1}) of Cd or As in rice roots, respectively ($n = 6$; $^*P < 0.05$, $^{**}P < 0.01$)

Parameters	Soil pH	Pore-water Si	Pore-water P	Cd in roots	As in roots
Pore-water Cd	-0.993^{**}	-0.953^{**}	-0.876^{*}	0.918^{**}	
Pore-water As	0.932^{**}	0.983^{**}	0.911^{*}		0.950^{**}

adsorption or precipitation (Wang et al. 2019). The strongly alkaline nature gives steel slag a high capacity to increase pH in acid environment (Navarro et al. 2010), likely leading to the precipitation of soil Cd as cadmium hydroxides. Besides the pH, the amount of high Si and P in steel slag enhanced the concentrations of Si and P in pore-water (Fig. 1). Wang et al. (2018a) also found that steel slag application increased the extractable Si and P in paddy soils. Improving Si and P nutrition could decrease the solubility and availability of Cd in soil by inducing the formation of Cd–Si co-precipitation and insoluble Cd phosphate (Sarwar et al. 2010). Lu et al. (2014) reported that Si additions restrained Cd uptake by crops, largely due to the alteration in soil chemistry, e.g., increasing soil pH and Cd adsorption, and reducing Cd competitiveness for crop uptake. Similarly, previous researches revealed that steel slag amendment decreased the extractable soil Cd related to the improvement of pH and extractable Si in pot experiments (Ning et al. 2016; He et al. 2017). Nevertheless, it was observed that the soluble P raised by steel slag might be another factor stimulating soil Cd immobilization in the present experiment (Fig. 1 and Table 4). Altogether, the mechanisms underlying the alteration of soluble soil Cd caused by steel slag are likely the increases of soil pH and soluble Si and P in pore-water (Fig. 1), as confirmed by the regression analysis in this study (Table 4).

Regarding As, unlike Cd, the present regression analysis confirmed the positive relationships of pore-water As concentrations with soil pH and pore-water Si and P concentrations, after steel slag amendments (Table 4). Analogously, the biochar and compost amendments promoted the solubilization of As to pore-water associated with increased pH and soluble P in soil (Beesley et al. 2014). It was explained that the pH increases by soil amendments decreased the As adsorption on soil minerals, and phosphate competed with As for binding sites in soil (Fleming et al. 2013; Beesley et al. 2014). Further, Si additions increased the As concentrations in soil solution due to the Si competition for As adsorption sites of soil solids (Lee et al. 2014). In general, alkaline materials, such as lime, fly ash, and biochar, may be undesirable for remediating As-contaminated soils as they cause higher pH with higher leaching and mobility of As (Kumpiene et al. 2008, 2019). Consequently, steel slag enhanced the solubility of soil As possibly by a combined effects of increased soil pH and pore-water Si and P (Fig. 1).

Generally, the solubility and availability of heavy metals in soil are the primary factors affecting the uptake of these metals by plant roots (Zeng et al. 2011). In this study, steel slag amendments decreased Cd concentrations and increased As concentrations in rice tissues (roots, straw, and brown rice) (Table 2), and the regression analysis revealed strongly positive correlations of Cd and As in roots with their concentrations in pore-water, respectively (Table 4). In agreement with our results, the declined concentrations of Cd in rice plants upon the addition of steel slag were observed, causing the decrease in extractable soil Cd (Ning et al. 2016; He et al. 2017). León-Romero et al. (2018) found that steel slag application promoted As uptake by roots and stems of *Arabidopsis thaliana* in pot experiments, regarding the possible reasons that were the alterations in As absorption and phosphate in soil. However, besides the pore-water P, the present results implied that soil pH and pore-water Si might generate evident impacts on the increases in the solubility and accumulation of As induced by steel slag amendments (Table 4). In addition, steel slag amendments decreased Cd but enhanced As in iron plaque, though the formation of iron plaque was not markedly increased by the amendments (Fig. 2). It is noted that iron plaque uptake of metals depends much on the bioavailability of metals in plant rhizosphere and the amounts of iron plaque, despite the wide recognition that iron plaque has a capacity to retain metals (Cheng et al. 2014). Similar to our results, Zheng et al. (2012, 2015) reported that the biochar amendments induced less Cd in iron plaque compared with the controls, owing to the decreased Cd in pore-water, thereby generating less transfer to the root surfaces from soil solutions. In contrast, the elevated pore-water As by steel slag amendments may have affected As onto the iron plaque and its concentrations in the plaque (Figs. 1 and 2). However, Yu et al. (2017) found that iron-based amendments reduced As dissolution in rhizosphere soils and increased iron plaques, contributing to decrease As concentrations in rice plants in pot experiments. Rice As accumulation varied among soil amendment systems, likely owing to the differences of the amendments used and their effects on As bioavailability in soil, iron plaque formation, and As mobility within rice plants. Thus, the formation of iron plaque and the plaque effects on rice As accumulation varied possibly. Furthermore, Cd translocation from roots to aerial parts was restrained by steel slag additions (Table 2). One possible explanation is the increases of Si concentrations and its co-precipitation with Cd in rice tissues (Ning et al. 2016). However, no significant alteration of As translocation by steel slag amendments may be attributed to the complicated pathways of As transport within rice plants, involving mechanisms of arsenate reduction, arsenite transport, As complexation and sequestration, As methylation, and As alteration in xylem and phloem (Zhao et al. 2010). More detailed investigations are required to understand the role of steel slag for the transfer process of As in crop plants.

Crop growth is also an important factor for evaluating the efficiency of soil amendment in agriculture lands (Kumpiene et al. 2019). Here steel slag amendments clearly improved the biomass of rice plants (roots, straw, and grains) and the activities of root antioxidant enzymes (Table 3). As reported, steel slag additions raised soil pH and resulted in a more neutral soil environment form the acidic one, benefiting rice growth (He et al. 2016). Besides, the increased solubility of nutrients like Si and P by steel slag additions contributed to rice biomass (Wang et al. 2015a, 2018a) and root antioxidant enzyme activities (Sarwar et al. 2010). Overall, the application of steel slag can be beneficial for grain production and Cd (not As) reduction in rice under field condition.

Conclusions

This study demonstrated that steel slag amendment effectively increased rice production and decreased the concentrations of Cd in rice tissues (roots, straw, and brown rice) and in iron plaque, though enhanced those of As, in a historically co-contaminated paddy field with Cd and As. The possible mechanism behind the effects of steel slag on Cd and As in rice is likely the slag decreasing soluble Cd and increasing soluble As in pore-water, related to significantly increased soil pH and soluble Si and P in pore-water, and suppressing Cd translocation from roots to aerial parts. Besides, the increments in soil pH and soluble nutrients (Si and P) cloud conduce to the improvement of rice yield and root antioxidant enzymes. These results suggest that steel slag amendment represents a favorable potential for the remediation of Cd-contaminated paddy soils, but seems undesirable for Cd and As co-contamination. Further investigations of field trials in multi-element contaminated sites are needed to evaluate the long-term effectiveness of the amendment.

Funding information This work was supported by the National Key Research and Development Program of China (2018YFD0800700), the National Natural Science Foundation of China (41501355), the Scientific Research Foundation for High-level Talents of West Anhui University (WGKQ201702007), the National Student's Project for Innovation and Entrepreneurship Training Program of China (201810376026), and the Borrowing Transfer to Supplement Foundation of Hefei (J2019D04). We sincerely thank Prof. Zhihong Ye (Sun Yat-sen University, China) for help in the improvement of this paper.

References

Beesley L, Moreno-Jiménez E, Clemente R, Lepp N, Dickinson N (2010) Mobility of arsenic, cadmium and zinc in a multi-element contaminated soil profile assessed by *in situ* soil pore water sampling, column leaching and sequential extraction. Environ Pollut 158:155–160

Beesley L, Inneh OS, Norton GJ, Moreno-Jimenez E, Pardo T, Clemente R, Dawson JJC (2014) Assessing the influence of compost and biochar amendments on the mobility and toxicity of metals and arsenic in a naturally contaminated mine soil. Environ Pollut 186:195–202

Cheng H, Wang MY, Wong MH, Ye ZH (2014) Does radial oxygen loss and iron plaque formation on roots alter Cd and Pb uptake and distribution in rice plant tissues? Plant Soil 375:137–148

Concas S, Ardau C, Di Bonito M, Lattanzi P, Vacca A (2015) Field sampling of soil pore water to evaluate the mobile fraction of trace elements in the Iglesiente area (SW Sardinia, Italy). J Geochem Explor 158:82–94

Fleming M, Tai YP, Zhuang P, McBride MB (2013) Extractability and bioavailability of Pb and as in historically contaminated orchard soil: effects of compost amendments. Environ Pollut 177:90–97

Guo JL, Bao YP, Wang M (2018) Steel slag in China: treatment, recycling, and management. Waste Manag 78:318–330

He HD, Tam NYN, Qiu RL, Yao AJ, Ye ZH (2016) Effects of alkaline and bioorganic amendments on cadmium, lead, zinc, and nutrient accumulation in brown rice and grain yield in acidic paddy fields contaminated with a mixture of heavy metals. Environ Sci Pollut Res 23:23551–23560

He HD, Tam NFY, Yao AJ, Qiu RL, Li WC, Ye ZH (2017) Growth and cd uptake by rice (*Oryza sativa*) in acidic and cd-contaminated paddy soils amended with steel slag. Chemosphere 189:247–254

Hu ZH, Zhuo F, Jing SH, Li X, Yan TX, Lei LL, Lu RR, Zhang XF, Jing YX (2019) Combined application of arbuscular mycorrhizal fungi and steel slag improves plant growth and reduces Cd, Pb accumulation in *Zea mays*. Int J Phytoremediat 21:857–865

Irshad MK, Noman A, Alhaithloul HAS, Adeel M, Rui YK, Shah T, Zhu SH, Shang JY (2020) Goethite-modified biochar ameliorates the growth of rice (*Oryza sativa* L.) plants by suppressing Cd and As-induced oxidative stress in Cd and As co-contaminated paddy soil. Sci Total Environ 717:137086

Kidd PS, Domínguez-Rodríguez MJ, Díez J, Monterroso C (2007) Bioavailability and plant accumulation of heavy metals and phosphorus in agricultural soils amended by long-term application of sewage sludge. Chemosphere 66:1458–1467

Kim KR, Kim JG, Park JS, Kim MS, Owens G, Youn GH, Lee JS (2012) Immobilizer-assisted management of metal-contaminated agricultural soils for safer food production. J Environ Manag 102:88–95

Kumpiene J, Antelo J, Brännvall E, Carabante I, Ek K, Komarek M, Söderberg C, Wårell L (2019) In situ chemical stabilization of trace element-contaminated soil – field demonstrations and barriers to transition from laboratory to the field – a review. Appl Geochem 100:335–351

Kumpiene J, Lagerkvist A, Maurice C (2008) Stabilization of as, Cr, cu, Pb and Zn in soil using amendments – a review. Waste Manag 28:215–225

Lee CH, Huang HH, Syu CH, Lin TH, Lee DY (2014) Increase of as release and phytotoxicity to rice seedlings in as-contaminated paddy soils by Si fertilizer application. J Hazard Mater 276:253–261

León-Romero MA, Soto-Ríos PC, Nomura M, Nishimura O (2018) Effect of steel slag to improve soil quality of tsunami-impacted land while reducing the risk of heavy metal bioaccumulation. Water Air Soil Pollut 229:12

Liu WJ, Zhu YG, Smith FA, Smith SE (2004) Do phosphorus nutrition and iron plaque alter arsenate (as) uptake by rice seedlings in hydroponic culture? New Phytol 162:481–488

Liu HJ, Li XP, Han XR, Liu YF, Lu JJ (2013) Effects of Fe-cd interaction on the lipid peroxidation and antioxidative enzyme activities of rice. Chin J Appl Ecol 24:2179–2185 **(in Chinese)**

Lu HP, Zhuang P, Li ZA, Tai YP, Zou B, Li YW, McBride MB (2014) Contrasting effects of silicates on cadmium uptake by three dicotyledonous crops grown in contaminated soil. Environ Sci Pollut Res 21:9921–9930

Moreno-Jiménez E, Beesley L, Lepp NW, Dickinson NM, Hartley W, Clemente R (2011) Field sampling of soil pore water to evaluate trace element mobility and associated environmental risk. Environ Pollut 159:3078–3085

Navarro C, Díaz M, Villa-García MA (2010) Physico-chemical characterization of steel slag. Study of its behavior under simulated environmental conditions. Environ Sci Technol 44:5383–5388

Nejad ZD, Kim JK, Jung MC (2017) Reclamation of arsenic contaminated soils around mining site using solidification/stabilization combined with revegetation. Geosci J 21:385–396

Ning DF, Liang YC, Liu ZD, Xiao JF, Duan AW (2016) Impacts of steel-slag-based silicate fertilizer on soil acidity and silicon availability and metals-immobilization in a paddy soil. PLoS One 11:e0168163

Otte ML, Rozema J, Koster L, Haarsma MS, Broekman RA (1989) Iron plaque on roots of *Aster tripolium* L.: interaction with zinc uptake. New Phytol 111:309–317

Rai PK, Lee SS, Zhang M, Tsang YF, Kim KH (2019) Heavy metals in food crops: health risks, fate, mechanisms, and management. Environ Int 125:365–385

Raj D, Maiti SK (2020) Sources, bioaccumulation, health risks and remediation of potentially toxic metal (loid) s (as, cd, Cr, Pb and hg): an epitomised review. Environ Monit Assess 192:108

Sarwar N, Saifullah MSS, Zia MH, Naeem A, Bibi S, Farid G (2010) Role of mineral nutrition in minimizing cadmium accumulation by plants. J Sci Food Agri 90:925–937

Wang C, Wang WQ, Sardans J, Singla A, Zeng CS, Lai DFY, Peñuelas J (2018b) Effects of steel slag and biochar amendments on CO_2, CH_4, and N_2O flux, and rice productivity in a subtropical Chinese paddy field. Environ Geochem Health 41:1419–1431

Wang P, Chen HP, Kopittke PM, Zhao FJ (2019) Cadmium contamination in agricultural soils of China and the impact on food safety. Environ Pollut 249:1038–1048

Wang WQ, Sardans J, Lai DYF, Wang C, Zeng CS, Tong C, Liang Y, Peñuelas J (2015a) Effects of steel slag application on greenhouse gas emissions and crop yield over multiple growing seasons in a subtropical paddy field in China. Field Crop Res 171:146–156

Wang WQ, Sardans J, Wang C, Zeng CS, Tong C, Bartrons M, Peñuelas J (2018a) Steel slag amendment increases nutrient availability and rice yield in a subtropical paddy field in China. Expl Agric 54:842–856

Wang X, Yao HX, Wong MH, Ye ZH (2013) Dynamic changes in radial oxygen loss and iron plaque formation and their effects on cd and as accumulation in rice (*Oryza sativa* L.). Environ Geochem Health 35:779–788

Wang X, Tang NFY, He HD, Ye ZH (2015b) The role of root anatomy, organic acids and iron plaque on mercury accumulation in rice. Plant Soil 394:301–313

Yu HY, Wang XQ, Li FB, Li B, Liu CP, Wang Q, Lei J (2017) Arsenic mobility and bioavailability in paddy soil under iron compound amendments at different growth stages of rice. Environ Pollut 224:136–147

Zeng F, Ali S, Zhang HT, Ouyang YN, Qiu BY, Wu FB, Zhang GP (2011) The influence of pH and organic matter content in paddy soil on heavy metal availability and their uptake by rice plants. Environ Pollut 159:84–91

Zhao FJ, McGrath SP, Meharg AA (2010) Arsenic as a food chain contaminant: mechanisms of plant uptake and metabolism and mitigation strategies. Ann Rev Plant Biol 61:535–559

Zheng RL, Cai C, Liang JH, Huang Q, Chen Z, Huang YZ, Arp HPH, Sun GX (2012) The effects of biochars from rice residue on the formation of iron plaque and the accumulation of cd, Zn, Pb, as in rice (*Oryza sativa* L.) seedlings. Chemosphere 89:856–862

Zheng RL, Chen Z, Cai C, Tie BQ, Liu XL, Reid BJ, Huang Q, Lei M, Sun GX, Baltrėnaitė E (2015) Mitigating heavy metal accumulation into rice (*Oryza sativa* L.) using biochar amendment - a field experiment in Hunan, China. Environ Sci Pollut Res 22:11097–11108