

Research Article

Cadmium Accumulation Characteristic in a Soil-rice System in Zhejiang Province, China

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Abstract: Cadmium (Cd) pollution of rice grain or soil is of increasing concern. This study investigates the concentrations of Cd in soil-rice system in parts area of Zhejiang province and analysis the effects of soil pH and organic matter content on the available Cd concentration in soil and the Cd accumulation of rice grain. The results showed that Cd concentrations in 90 rice grain samples range from 0.01 to 0.31 mg/kg, with an average of 0.1 mg/kg. There is high Cd accumulation in soil that 40% of soil samples were polluted by Cd. The available Cd concentrations in soils had a range of 0.01-0.25 mg/kg, with an average of 0.10 mg/kg. The total Cd concentrations in soil were not the uniquely determined factor for the Cd accumulation of rice grain, Cd concentrations in rice grain were also significantly positive correlated with the available Cd concentrations in soil. The soil pH value and SOM also negatively correlated with available Cd concentrations in soils and Cd concentration in corresponding rice grain. The results indicated the variations of Cd concentration in rice grain were related to Cd concentrations in corresponding soils. Also, soil pH and organic matter content influence the Cd concentrations in soils and affected Cd absorption in rice.

Keywords: Cadmium, organic matter content, soil-rice system, soil pH, Zhejiang province

INTRODUCTION

Cadmium (Cd) is regarded as one of the nonessential and toxic trace element in the environment and with no known benefit to plants, animals or humans (Järup and Akesson, 2009; Wang *et al.*, 2013a). Due to:

- Atmospheric fallout from industrial and urban activities
- Application of sewage sludge and manures
- Widespread use of chemical fertilizers, Cd is widely released into agricultural soils, air and water (Liu *et al.*, 2007; Yang *et al.*, 2006; Mar *et al.*, 2012; Wang *et al.*, 2013b)

Thus, the contamination and accumulations of Cd in grains and vegetables have aroused wide attention by the public (Fang *et al.*, 2014; Kosolsaksakul *et al.*, 2014). Paddy rice is one of the main food sources in the world, while Cd is easily taken up and accumulated by rice plant and negatively influenced plant growth and development (Liu *et al.*, 2007). The contamination of paddy rice with Cd is invisible and irreversible, with potentially devastating consequences for safe production of rice crops and human health (Liu *et al.*, 2007; Grant and Sheppard, 2008). Itai-itai disease was

the infamous and terrible case of Cd toxicity, which caused by the daily ingestion of Cd-contaminated rice that emerged in Japan in the middle of the 20th century (Tsukahara *et al.*, 2003). In recent year, the Cd pollution in the edible parts of rice grains and rice products has become increasingly serious in some parts of China (Fang *et al.*, 2014; Zhu *et al.*, 2008; Huang *et al.*, 2009; Hang *et al.*, 2009). In China, there are about 5.0×10^7 kg of rice is polluted by Cd (Wang, 2002). Zhen *et al.* (2008) discovered that 10% of commercially available rice grain in China had excessive levels of Cd. Soils are the main source of Cd available to plants, which can become the main source of Cd in plant-derived foods in human diets (Liu *et al.*, 2007). In the recent past, the issue of Cd pollution in agricultural soils has become more serious (Yang *et al.*, 2006; Herawati *et al.*, 2000). Cd accumulate and migrate in soil is easily taken up by plants and it can accumulate in the human body through the food chain and therefore pose a potential risk for both animals and humans (Liu *et al.*, 2007; Singh *et al.*, 2011). The pollution of Cd in agricultural soils has seriously affect agricultural environment quality and food safety and also become an important constraining factor of pollution-free rice in China.

In the recent past, the issue of Cd pollution in Chinese agricultural systems has become more serious.

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Cd pollution of rice grain or soil has become a very hot topic for scientists in recent years. Researchers have carried out many surveys and experiments about the sources of Cd pollution and other issues in agricultural soil or plants. However, there are few surveys and little information relating to the migration and accumulation characteristics of Cd trace elements in soil-rice systems, or the factors that influence them. Paddy rice is the main cereal in China. Zhejiang is one of main rice-growing area. Therefore, the main objectives of this study were to determine the degree of Cd pollution in rice grain and the corresponding soils, so as to clarify the factors that influence Cd accumulation in rice grain. The results of our study may provide useful information about the mechanisms by which Cd accumulate in soil to support the safe production of rice grain in China in the future. The findings from this study may also establish a foundation for safe agricultural production and improved human health.

MATERIALS AND METHODS

Sample collection: A total of 50 pairs of rice grain and soil samples were collected from a main rice production area-Zhejiang province, China (23°30'-28°22'N, 115°50'-120°40'E). Zhejiang is located in the southern part of the Yangtze River Delta on the southeast coast of China. We designed a suitable route and chose the sampling areas (1-15 Hangzhou city, 16-30 Jiaxing city, 31-45 Taizhou city, 46-60 Fuyang city, 61-75 Quzhou city, 76-90 Ningbo city) and then went to the rice planting areas that were generally located between two towns. Samples were collected from large, thriving, representative paddy fields in the mature stage of rice growth. Between five and ten mature rice panicles were harvested from each paddy field. Soil samples of approximately 300 g were collected at the top (0-20 cm) of the soil profile from the same paddy fields. All the samples were collected in October, 2013.

Sample pretreatment: Soil samples were air-dried indoors at room temperature. They were crushed with a wooden stick, then passed through a 2.5 mm nylon sieve to remove grit and plant residues and then were ground finely with an agate mortar. They were screened through a 100-mesh nylon sieve and a subsample of 50 g was bagged and stored in a dry place for further analysis.

The samples of rice grain were rinsed with tap water, washed with distilled water and dried in the oven. They were then shelled with a rice husking machine and ground to powder for further analysis.

Sample analysis methods: Soil pH was measured using the glass electrode method of Chaturvedi and Sankar (2006), with a water to soil ratio of 2.5:1; Soil Organic Matter (SOM) content was determined by the chromic acid titration method (Ryan *et al.*, 2001); The available-Cd content was determined by the EDTA-Na₂

extraction method (Li *et al.*, 2003). Total soil Cd was determined by graphite furnace atomic absorption spectrometry (AA-7000) with mixed acid digestion (HNO₃-HF). The concentrations of Cd were measured with graphite furnace atomic absorption spectrometry (AA-7000).

Total soil Cd was determined by graphite furnace atomic absorption spectrometry (AA-7000) with mixed acid digestion (HNO₃-HF). About 0.2 g of each of the above-mentioned soil samples was weighed and placed into digestion vessels. High purity HNO₃ (6 mL) and high purity HF (2 mL) were added to the vessels, which were then placed into a ventilation cabinet for 2-3 h. The samples were then digested in a microwave digester (CEM-MARS). Similarly, The Cd concentrations of rice samples were determined by AA-7000 following HNO₃-H₂O₂ digestion procedures. About 0.3 g of the rice grain samples were weighed and placed into digestion vessels. The vessels were kept in a ventilation cabinet for 2-3 h after 6 mL high purity HNO₃ and 0.5 mL H₂O₂ was added and then they were placed into the microwave digester (CEM-MARS) for digestion. After digestion, the mixture was reduced to 1 mL at high temperature. The mixture solutions were then diluted into 25 mL volumetric flasks with ultra-pure water and obtained the clarification samples, then stored in the refrigerator at 4°C for further analysis.

Statistical analysis: Statistical analyses were done using Microsoft Excel and SPSS 18.0 software, using significance level of $p < 0.05$ and $p < 0.01$.

RESULTS

Cd concentration in rice grain: The concentrations of Cd (mg/kg, dry weight basis) in rice are presented in Fig. 1 and Table 1. The Cd concentrations in the rice grain were very variable, ranging from 0.01 to 0.31 mg/kg, with an average of 0.1 mg/kg. There are about 23 samples that Cd concentrations in the rice grain exceeded the National Food Sanitary Standards of China (GB2715-2005) ($Cd \leq 0.2$ mg/kg). Results show that 25.5% of the 90 soil samples were polluted by Cd. Some rice grains from taizhou, fuyang and ningbo were seriously polluted. These results indicate that rice grain in Zhejiang was in some degree polluted by Cd.

Cd concentration in soils: Information about Cd contamination of soil from some areas of Zhejiang Province is presented in Fig. 2 and Table 1. The total Cd concentrations in soils ranged from 0.06 to 0.67 mg/kg, with an average of 0.24 mg/kg. Meanwhile, the available Cd concentrations in soils varied between 0.01 and 0.25 mg/kg, with an average of 0.10 mg/kg. Comparison with the Chinese National Environmental Quality Standards for soil (GB15618-1995) showed that the local soil background levels for Cd was exceeded in 40% of the 90 soil samples, which indicates that the wide spread of Cd accumulation in the soils sample from Zhejiang Province.

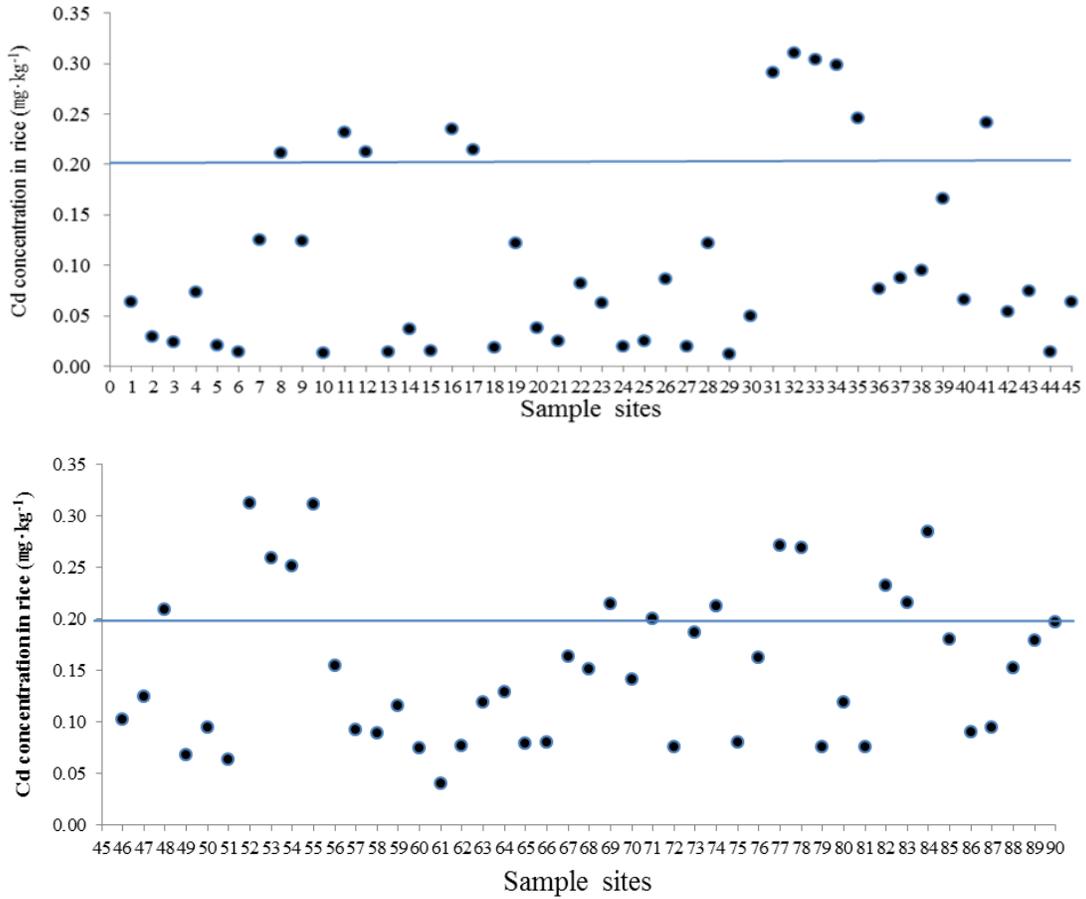
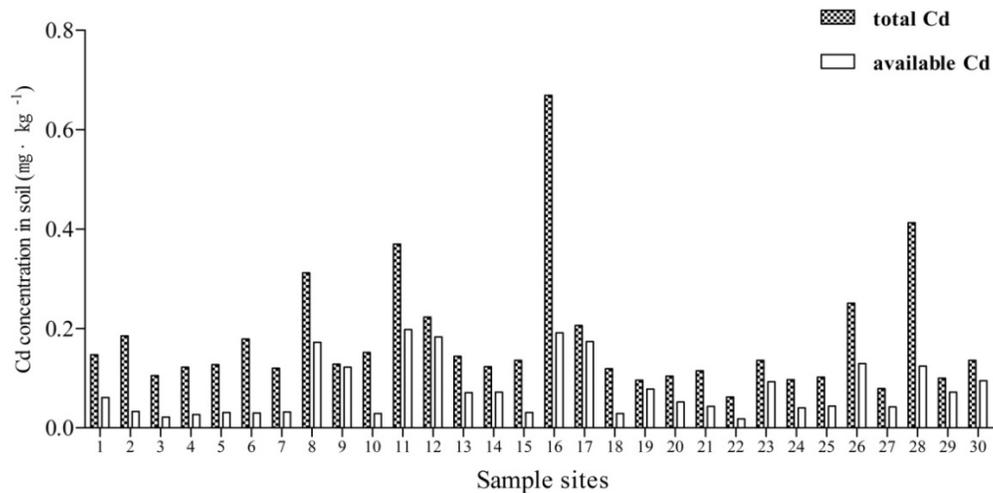


Fig. 1: The concentrations of Cd in rice grain of the sampling areas in Zhejiang; Serial number of sampling points: 1-15 Hangzhou city, 16-30 Jiaxing city, 31-45 Taizhou city, 46-60 Fuyang city, 61-75 Quzhou city, 76-90 Ningbo city

Table 1: The concentrations of Cd in soil and rice grain, soil pH value and soil organic matter from Zhejiang

	Total Cd concentration in soil	Available Cd concentration in soil	Cd concentration in rice	Ph value	Soil organic matter
Number	90	90	90	90	90
Max	0.67	0.25	0.31	6.98	45.40
Min	0.06	0.01	0.01	3.99	12.38
Overall mean	0.24	0.11	0.13	5.95	27.51
Limit (\leq)	0.20		0.20		
The percentage of excessive	40%		25.50%		



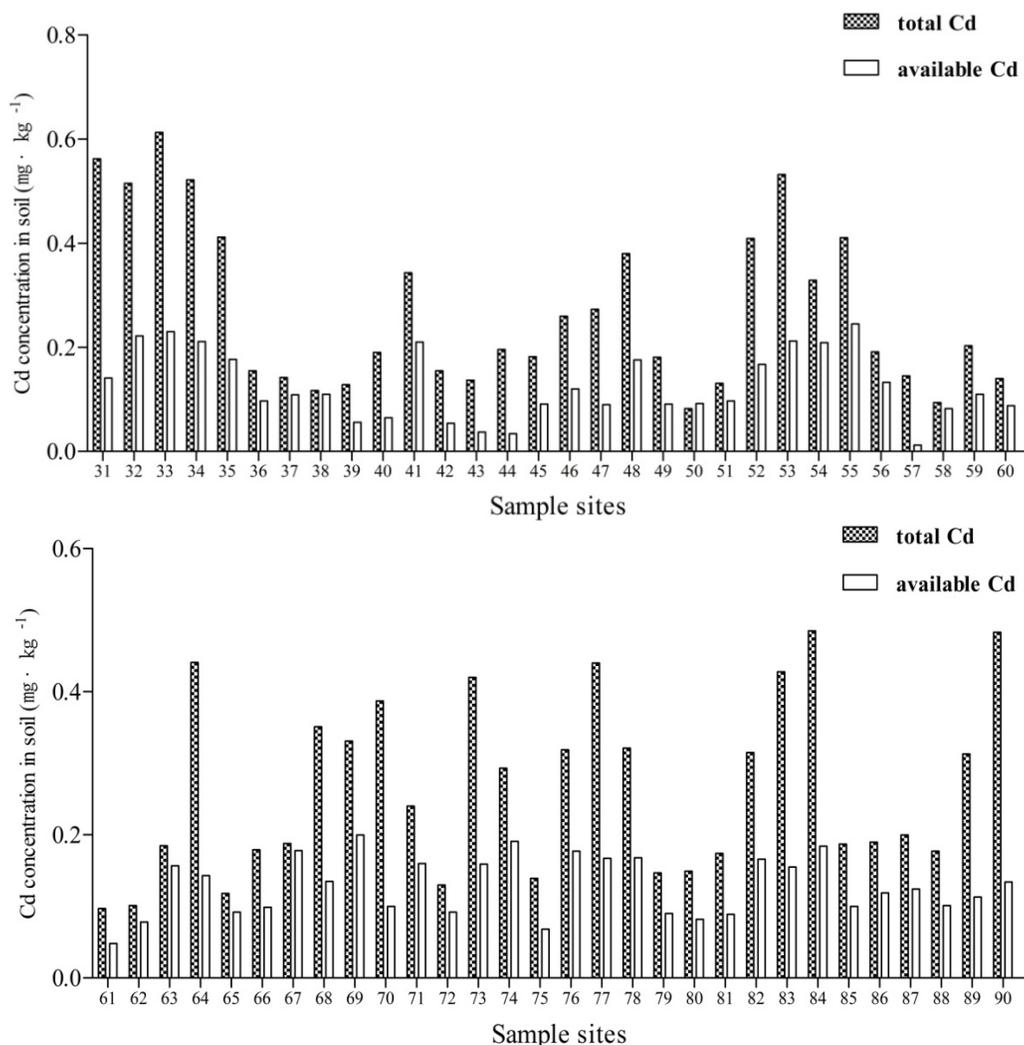


Fig. 2: The concentrations of Cd in soil of the sampling areas in Zhejiang; Serial number of sampling points: 1-15 Hangzhou city, 16-30 Jiaxing city, 31-45 Taizhou city, 46-60 Fuyang city, 61 -75 Quzhou city, 76-90 Ningbo city

Relationship between Cd contamination in soil and rice grain:

The Correlation between Cd contamination in soil and Cd concentrations in rice grain samples is showed in Fig. 3 and Table 2. Correlation analysis showed that the total concentrations of Cd in the soil and that rice grain samples was significantly positive correlated ($r = 0.721, p < 0.01$). There was more significant positive correlation ($p < 0.01$) between available Cd concentrations in soils and Cd concentrations in rice grain ($r = 0.880$) (Fig. 4). In some sample sites where the soil was contaminated with Cd, the available Cd concentrations in soil were higher and the Cd concentrations in the corresponding rice grain exceeded the limit values. We can therefore conclude that the concentration of Cd in rice grain was positively spatially correlated with total Cd and available Cd concentrations in corresponding soils. However, the Cd accumulation in rice grain had stronger correlations with the available Cd concentrations in soil, which

suggests that soil properties may potential influence Cd accumulation in rice grain.

Determination of pH and SOM in the soil:

Determination of the pH and the SOM in the soil: In order to understand the potential effects of parts soil properties on the availability of Cd to rice, soil pH values and SOM were determined. Information on soil pH values and SOM of all the soil samples is shown in Table 2. Soil pH values in the samples from Zhejiang Province ranged from 3.99-6.98 and had an average value of 5.95. The average SOM in the soil samples was 27.51 mg/g and ranged from 12.38 to 45.40 mg/g. The relationships between the soil pH and Cd concentrations in the rice grain and soil are shown in Table 3. There was a significant negative correlation between the soil pH values and available total Cd concentrations in soil ($r = -0.820, p < 0.01$) (Fig. 5) and the correlation coefficient between soil pH and Cd

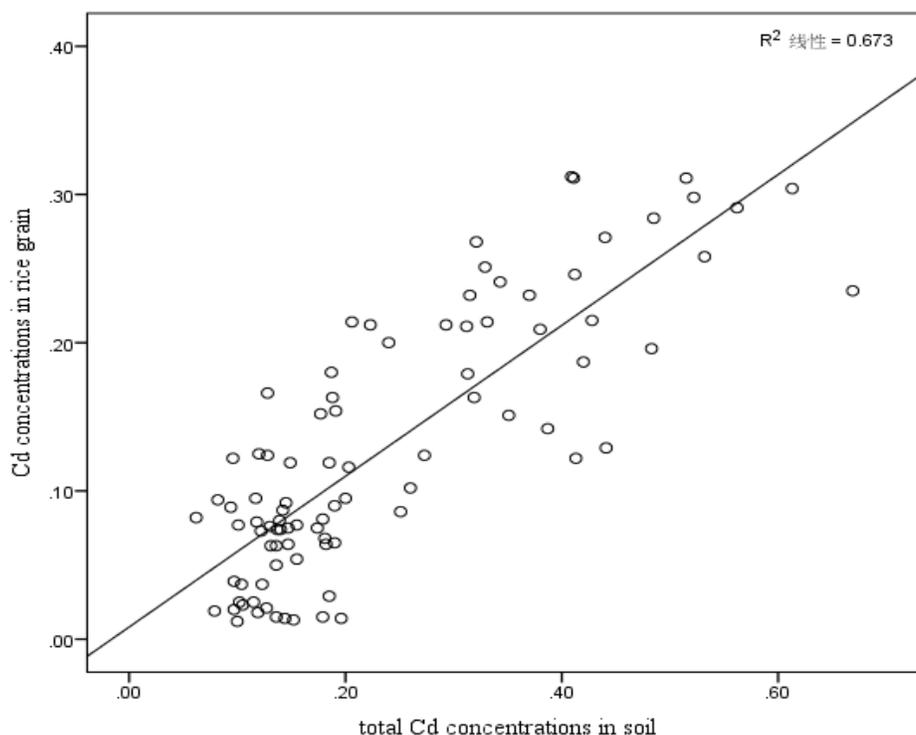


Fig. 3: Correlation between total Cd concentrations in soil and Cd concentration in rice

Table 2: The soil pH and SOM content of the samples in Zhejiang

Location	Ph	Som	Location	Ph	Som	Location	Som	Ph	Location	Ph	Som
1	6.41	31.64	26	6.41	24.39	51	6.70	26.39	76	5.49	12.38
2	6.67	34.02	27	6.81	39.90	52	4.94	17.15	77	4.43	16.51
3	6.63	43.28	28	5.37	24.76	53	4.68	16.15	78	5.24	21.32
4	6.83	42.65	29	6.69	33.02	54	5.06	18.26	79	5.81	33.02
5	6.83	39.90	30	6.20	30.27	55	5.22	13.76	80	6.20	34.27
6	6.73	34.39	31	4.84	20.01	56	6.19	20.21	81	6.24	32.27
7	6.98	38.52	32	4.97	13.75	57	6.86	37.52	82	4.51	15.82
8	5.18	21.52	33	5.03	15.13	58	6.43	20.64	83	4.89	17.57
9	5.06	24.58	34	4.96	16.78	59	6.26	20.64	84	5.04	19.26
10	6.62	40.53	35	4.75	18.25	60	6.30	27.52	85	5.79	20.64
11	4.80	20.27	36	6.21	21.25	61	6.20	34.52	86	5.39	27.52
12	5.12	23.39	37	6.29	24.02	62	6.41	28.89	87	5.54	22.38
13	6.76	38.27	38	6.26	28.52	63	6.31	20.27	88	5.99	28.01
14	6.91	40.78	39	6.41	34.76	64	5.87	22.88	89	5.78	21.32
15	6.80	39.9	40	6.67	41.27	65	6.89	39.90	90	5.79	20.13
16	5.62	12.38	41	3.99	17.88	66	6.26	45.40			
17	4.94	19.26	42	6.63	34.39	67	6.43	20.14			
18	6.55	40.78	43	6.54	38.27	68	6.26	32.33			
19	6.00	45.4	44	6.69	26.14	69	4.94	24.64			
20	6.21	40.9	45	6.26	26.51	70	5.45	30.95			
21	6.43	38.52	46	5.68	24.02	71	5.86	16.51			
22	6.19	44.02	47	5.86	28.15	72	6.43	20.38			
23	6.06	30.15	48	5.61	18.39	73	5.66	18.57			
24	6.43	36.39	49	6.80	26.14	74	5.69	20.45			
25	6.30	34.39	50	6.42	28.01	75	6.64	29.95			

Table 3: Correlation among pH and SOM with Cd concentration in soil and rice

	Total Cd concentration in soil	Available Cd concentration in soil	Cd concentration in rice	pH value	Soil Organic Matter (SOM)
Total Cd concentration in soil	1				
Available Cd concentration in soil	0.774**	1			
Cd concentration in rice	0.721**	0.880**	1		
pH value	-0.734**	-0.82**	-0.843**	1	
Soil organic matter	-0.704**	-0.848**	-0.764**	0.713	1

** : Correlation is significant at the 0.01 level

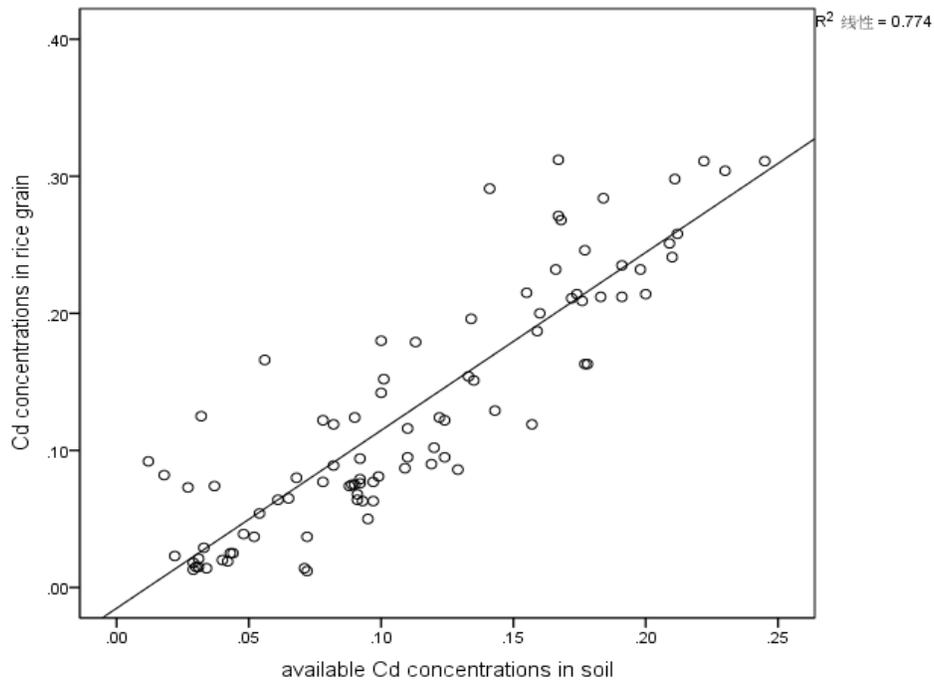


Fig. 4: Correlation between available Cd concentrations in soil and Cd concentration in rice

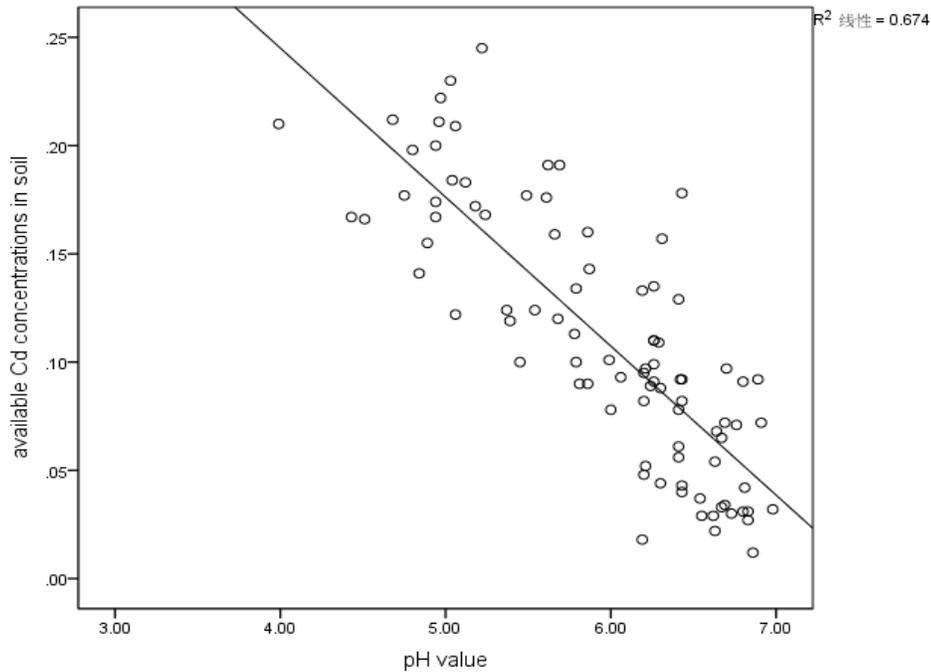


Fig. 5: Correlation between available Cd concentrations in soil and soil pH value

concentrations in rice grain was - 0.843 ($p < 0.01$) (Table 3). Generally, rice grain samples and the corresponding soils had high Cd concentrations that soil pH values were less than 5.0. In general, as the pH value decreased, the absorption of Cd in rice grain increased.

The relationships between SOM and Cd concentrations in rice grain and soil are shown in Table 3. The total Cd and available Cd concentrations in the

soil were significantly negative correlated with SOM, with correlation coefficients of -0.704 ($p < 0.01$) and -0.848 ($p < 0.01$) (Table 3 and Fig. 6). Correlation analysis between SOM and the accumulation of Cd in rice grain showed that there was a significant negative correlation between the Cd concentration and SOM ($r = -0.764$, $p < 0.01$). We can conclude that SOM can

influence the concentrations of Cd in rice grain to a certain extent, as demonstrated by the influence Cd concentrations soil.

DISCUSSION

Cd pollution in soil and rice grains: When the samples from the different areas are compared, there is a regional pattern in Cd pollution. Cd pollution of rice grains was more serious in taizhou, fuyang and ningbo than in other places. Cd pollution in agricultural soils is mainly due to human activities, irrigation with industrial wastewater, atmospheric fallout from industrial (Facchinelli *et al.*, 2001; Martín *et al.*, 2006). Studies have shown that Cd concentrations in plants may related to that transport emissions (Bakirdere and Yaman, 2008; Chen *et al.*, 2010; Saeedi *et al.*, 2009). Air pollution, sewage water irrigation and pesticide spraying also can influence the concentrations of Cd in plants (Chary *et al.*, 2008; Huang *et al.*, 2007). The soil-rice pollution therefore may be related to human activities and industrial pollution, as was also suggested by other studies. For example, higher Cd concentrations in soil-crop systems around mines and factories than in less industrial areas (Kim *et al.*, 2008; Liu *et al.*, 2010; Zhuang *et al.*, 2009a, 2009b). Our results showed that the soil was not polluted by Cd, but that the concentrations of Cd exceeded the threshold values in rice grain; this may be related to sewage water irrigation to the soil surface, Cd deposition from the air and the variety of rice being cultivated (Chung *et al.*, 2011; Feng *et al.*, 2011; Luo *et al.*, 2011).

Relationship between Cd concentrations in soil and rice grains: There were significantly positive correlations exist between total Cd concentrations in soil and the Cd concentration in rice grain. Rogival *et al.* (2007) showed that the total concentrations of Cd in soil were not the sole determinant of the Cd accumulation in crops. There are many chemical forms of soil heavy metals (Aydinalpi and Marinova, 2003; Zalidis *et al.*, 1999), of which the dissolved and exchangeable states are easily absorbed by crops. In our results, we also found the Cd concentrations in rice grain were significantly positive correlated with the available Cd concentrations in soil. In general, the correlation between Cd accumulation in rice grain and the available Cd content in soil was stronger than the correlation between Cd accumulation in rice grain and the total Cd content in soil.

Effects of soil pH on Cd contamination in soil and rice grain: Soil pH is the factor that controls the ability of plants to absorb heavy metals and the speciation and the availability of heavy metals in soil (McBride, 2002; Singh and Myhr, 1998; Zhao *et al.*, 2010). Our study

demonstrate that soil pH influence Cd concentrations in soil and rice grain. Zhao *et al.* (2010) also showed that soil pH influenced Cd absorption by rice grain. Correlation analysis showed that there was a significant negative relationship between soil pH and available Cd concentrations in soil, the Cd concentrations of rice grain. This means that, to a certain degree, as the soil pH value increased, reduced the availability of Cd, the Cd concentrations in the rice grain decreased. Low pH may result in increased solubility and high availability of Cd for rice (Zhao *et al.*, 2010). In other words, Soil pH influenced the dissolution of Cd, particularly in acid paddy field (Wu *et al.*, 2010). Singh and Pandeya (1998) found, that as the soil pH value increased, the negative charge of soil colloids increased and the H⁺ ion competitiveness weakened, so that heavy metals that were at one point firmly bound together were transformed into insoluble salt forms, like hydroxide, carbonate or phosphate; this process would significantly reduce the availability of Cd.

Effects of SOM on Cd contamination in soil and rice grain: As well as soil pH, SOM has an important influence on the availability of Cd in soil. Singh and Pandeya (1998) showed that SOM could significantly reduce the availability of heavy metals in soil, thereby reducing the absorption of heavy metals in crops. Our results show that there were negative relationships between SOM and the available Cd concentrations in soil samples from the whole region. SOM was a factor that had an influence on available Cd in soil, then effect the absorption of Cd in rice grain. The effect of SOM on the availability of Cd could be due to lower solubility of Cd in soils and the availability of Cd decreased with increasing SOM (Huang *et al.*, 2007; Kashem and Singh, 2001). Other studies have indicated that SOM was the major influence on the exchange capacity of Cd and that exchange Cd accumulation in soil decreased as SOM decreased (Antoniadis *et al.*, 2008). Our results also suggest that the influence of SOM on paddy soil uptake of Cd was not only dependent on the Cd content, but also on the Cd components, such as the availability of Cd. This result also met with the reports by Ghosh *et al.* (2012) and Zeng *et al.* (2011).

The processes that control migration and transformations of Cd in soil-rice systems are extremely complex and there are many influencing factors. As well as the amount of Cd in soil, soil pH and SOM are major controls on Cd migration from soil to plants. The different rice types, as the rice genotypes also can influence the transfer and bioavailability of Cd in the soil-rice system (Huang *et al.*, 2007; Cheng *et al.*, 2006). Numerous studies also indicated that different soil types, soil electrical conductivity, clay content, nutrients, enzyme activity and other physical and

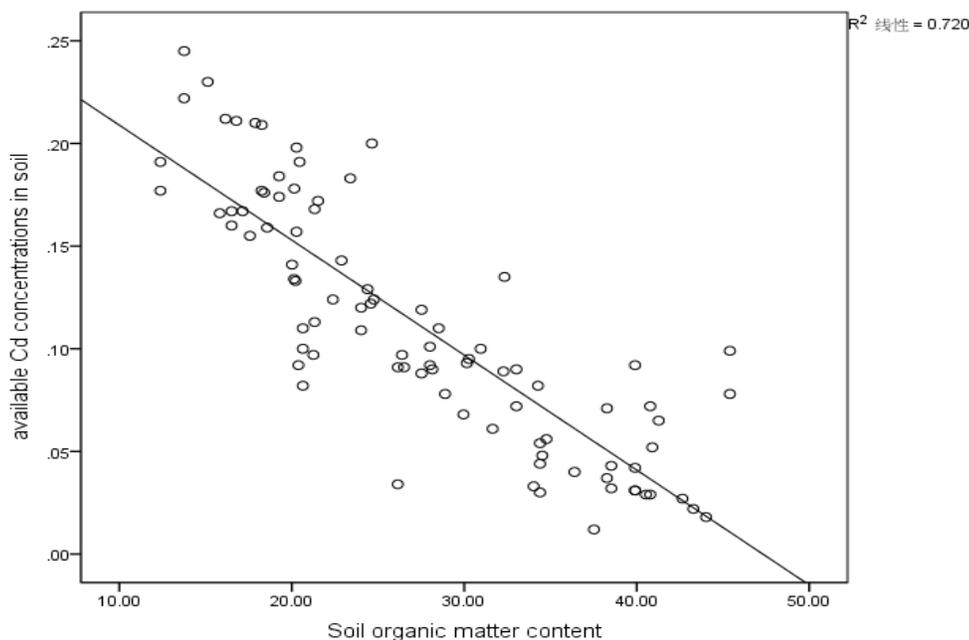


Fig. 6: Correlation between available Cd concentrations in soil and soil organic matter

chemical properties could directly influence the migration and the accumulation of Cd in soil-rice system (Adams *et al.*, 2004; Bañuelos and Ajwa, 1999; Jung and Thornton, 1997; Li *et al.*, 2007; Zhao *et al.*, 2010).

CONCLUSION

This experiment investigated the concentrations of Cd in soil-rice system in typical area of Zhejiang province. The results showed that a high Cd accumulation in soil-rice system that 25.5% of rice grain samples and 40% of soil samples were polluted by Cd. It was very likely the total Cd concentrations in soil were not the uniquely determined factor for the Cd accumulation of rice grain. Cd concentrations in rice grain were also significantly positive correlated with the available Cd concentrations in soil. The soil pH value and SOM also significantly influenced the available Cd concentration in soil and affected Cd absorption in rice. The results of this survey may provide basic information for the safety assessment of Cd in soil and rice, but also provides scientific basis for the safe production of rice.

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