



Distribution and availability of cadmium in profile and aggregates of a paddy soil with 30-year fertilization and its impact on Cd accumulation in rice plant[☆]

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ABSTRACT

The research was conducted to investigate the accumulation, distribution and availability of Cd in paddy soil and their relation to Cd in rice plants under 30-year fertilization regimes. Six treatments were involved in the study: control without fertilization (CK), chemical fertilizer (NPK), high nitrogen chemical fertilizer (HN), rice straw incorporation (ST), low and high dosage of manure fertilizer (LM and HM). Total and DTPA extractable concentration of Cd (T-Cd and DTPA-Cd) in bulk soils (20 cm topsoil), profiles (0–60 cm) and aggregates (>2, 1–2, 0.5–1, 0.25–0.5, 0.053–0.25 and <0.053 mm) were investigated. The Cd concentration in relevant rice plant (roots, stems, leaves, husks and grains) were also analyzed. Manure fertilizers caused T-Cd accumulation in bulk soil with a significant increase of 36.2% in LM and 81.2% in HM. Similar impacts of manure fertilizers were observed in DTPA-Cd in the bulk soil. Further, the HM generated a further accumulation in deeper soil layers, presenting a remarkable increase of T-Cd (28.3%–225%) in 10–40 cm and DTPA-Cd (116%–158%) in 10–30 cm profiles. Moreover, the continuous application of manure fertilizers enhanced the availability of Cd in all aggregate size classes with an increase of 17.3%–87.8% in DTPA-Cd. Organic fertilizers (LM, HM and ST) heightened the content of Cd (38.0%–152%) in all parts of rice plant. The accumulation of Cd in rice plants was directly affected by fertilization regimes and Cd availability in the 10–20 cm soil layers and 0.25–0.5 mm aggregates. In conclusion, long-term application of manures resulted in increasing availability of Cd in aggregates and in topsoil and subsoil layers, which accordingly enhanced the accumulation of Cd in rice plants.

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1. Introduction

Cadmium (Cd) is a hazardous heavy metal found in the environment. Anthropogenic activities have resulted in the release of a substantial amount of Cd into cropland and especially in paddy regions (Grant and Sheppard, 2008). Considering the high transfer ability of Cd in soil-rice system, the contamination risk of Cd in rice grain has been found all over the world, e.g. China and Southeast Asia (Zhao et al., 2015; Tang et al., 2016). Controlling the accumulation of Cd in rice grains have been a worldwide issue because of

the bioaccumulation of Cd in crops and its potential risks to human body through the food chain (Xu et al., 2015; Chen et al., 2017).

Previous studies indicated that Cd-bearing fertilizers, especially manures, may be one of the important sources for Cd entering into soil (Zhang et al., 2011; Cheng et al., 2013; Zhao et al., 2014). For example, livestock manures were observed to account for 55% of the total Cd inputs in agricultural soils of China (Luo et al., 2009). The effect of fertilization on Cd in soil has been related not only to the concentration and availability of Cd in bulk soils but also to the accumulation and distribution of Cd in soil profiles and aggregates (Acosta et al., 2011; Fan et al., 2012). Application of pig manures has been reported to significantly increase the Cd concentration in soils (Wu et al., 2012; Xu et al., 2015). It was also observed that Cd in subsoil layers increased significantly after continuous application of pig manures for 10 years (Xu et al., 2015). Furthermore, the

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availability of Cd may be strongly affected by soil aggregate size distribution because aggregates are the basic unit of soil construction and important place for chemical and biological reaction activities (Ilg et al., 2004; Fedotov et al., 2008). Increasing risk of Cd uptake by rice was also found in paddy soils fertilized with pig manure and green manure (Wang and Zhou, 2017). Long-term fertilization is likely to result in much more serious accumulation risks of heavy metals in soils and crops (Hejman et al., 2008; Zhao et al., 2015; Zhou et al., 2015). It is crucial to determine the distribution of Cd, and its availability, at the profile and aggregate scales, as affected by long-term fertilization regimes.

Therefore, the distribution and accumulation characteristics and availability of Cd in bulk paddy soil, profiles, aggregates and rice plants were analyzed using a 30-year fertilization experiment. The objective was to elucidate the effects of long-term fertilization on Cd availability in soil profiles and aggregates and relate their impacts on Cd accumulation in rice plant.

2. Materials and methods

2.1. Site and soil description

The long-term fertilization experiment was initiated in 1986 in Hunan Province, China. The site has a humid subtropical monsoon climate, an annual average temperature of 16.8 °C and a mean precipitation of 1358 mm. The tested soil was derived from modern river alluvial deposits and classified as alluvial sand soil. The pH of the tested soil is 5.6 on average, and it contains an average quality of soil organic carbon (SOC) 21.1 g kg⁻¹, total nitrogen (TN) 2.17 g kg⁻¹, total phosphorus (TP) 1.08 g kg⁻¹ and total potassium (TK) 22.3 g kg⁻¹.

2.2. Fertilization designs

The fertilization regimes had been applied and lasted for 30 years up to 2015, in which year the soil samples were collected. The fertilization treatments were: no fertilizer (CK), chemical fertilizer (NPK), high nitrogen chemical fertilizer (HN), rice straw incorporation (ST), low dosage of manure fertilizer (LM) and high dosage of manure fertilizer (HM). The dosage of chemical fertilizer (NPK) was determined according to the N, P and K supplying capacity of the soil and the absorption ability of plants in the supervised field. The source of N and K were urea fertilizer (46%) and potassium chloride fertilizer (60%), respectively. The P was applied in the form of calcium-magnesia phosphate fertilizer (12%). Thus, some other elements such as Ca, Mg and Si were also imported into soil coupled with P. High nitrogen chemical fertilizer (HN) involved further application of N, additional to the NPK treatment. The manure fertilizer used in the study was mainly pig and fowl manure, and the proportions of N in the low and high dosages of manure fertilizer were 30% and 60% of total N applied to the soil, respectively. The amount of ST was 3000 kg ha⁻¹. The LM, HM, and ST treatments were supplemented by chemical fertilizers. The details of fertilizer application are shown in Table 1.

2.3. Sampling

The bulk soil samples were collected from the 0–20 cm surface soil layer. To ensure the representation and uniformity of soil samples, a composite soil sample was obtained by thoroughly mixing 5 subsamples at each sampling site. Bulk soil samples were pre-treated by air drying and then ground to pass through 1 mm and 0.149 mm sieves for further analysis.

Profile samples were collected in the soil layers from depths of 0–60 cm and were divided into 6 layers with 10 cm per layer. A

Table 1

Field fertilization amount of tested soils (by purity nutrient content, kg/ha/a) (Di et al., 2014).

Treatments	Chemical fertilizer			Straw/Manure			Total amount		
	N	P	K	N	P	K	N	P	K
NPK	457	44	160	0	0	0	457	44	160
HN	757	44	160	0	0	0	757	44	160
ST	378	36	1	81	12	167	459	48	168
LM	313	30	61	147	56	106	460	86	167
HM	164	36	0	293	111	211	457	147	211

*Field fertilization amount (purity content) represented the average values between 2004 and 2011.

stainless steel earth drill with a scale line mark was used for sample collection. The profile samples were air dried and sieved to <0.149 mm for further analysis.

Water-stable aggregates were derived from 0 to 20 cm topsoil and separated into six size fractions by wet sieving (Elliott, 1986) as, >2, 1–2, 0.5–1, 0.25–0.5, 0.053–0.25 and <0.053 mm. Five sequential size sieves were used and placed in descending order from top to bottom. Before fractionation, the soil samples were submerged in distilled water for 10 min and then sieved vertically in distilled water by machine at a vertical reciprocating frequency of 35 r min⁻¹ over a period of 15 min. After separation, the samples in the sieve were collected into aluminum specimen boxes, and the samples <0.053 mm in the water were collected by settlement and centrifugation. All aggregate samples were then dried at 60–70 °C in an oven at constant temperature.

The plant samples were also collected together with the soil samples. The rice plants were collected in their entirety. After deactivation of enzymes at 105 °C, the rice plants were then divided into roots, stems, leaves, husks and grains. All the plant sample components were dried in oven at 60–70 °C and then ground into powder with a stainless steel pulverizer for further analysis.

2.4. Physical and chemical analysis

The soil physical and chemical properties (SOC, pH, and total N/P/K) were analyzed according to Lu (2000). The total Cd in the bulk soil, water-stable aggregates and soil profiles was solubilized by digestion of 0.5 g in 10 mL aqua regia (HCl: HNO₃ = 3:1, V/V; AR) and 2 mL HClO₄ (AR). The available Cd was extracted with a DTPA-TEA-CaCl₂ solution by shaking for 2 h at 180 r min⁻¹. This DTPA method is widely used and regarded as national standard protocols to investigate soil extractable Cd in China (Zhu et al., 2016). The Cd in rice plants was solubilized by digestion of 1.0 g in 10 mL HNO₃ (AR) and 2 mL HClO₄ (AR). All detected soil and plant samples were digested or extracted in two duplicate. Distilled deionized water was used for blank correction and the referenced soil (GBW07405) was used as certified reference material. Both of the blank and reference material were digested in the same pattern of soil samples.

All digestion and extraction solutions were finally analyzed via inductively coupled plasma optical emission spectrometry (ICP-OES, Varian, US), with a limit of 0.003 µg mL⁻¹ for Cd. A stock solution of Cd (1000 µg mL⁻¹, Merck) was used as an external standard by diluting it into 1 µg mL⁻¹. Moreover, all data were blank-corrected and the standard curve was checked every ten samples during the detection procedure.

2.5. Statistical analysis

All statistical analyses were performed via Excel 2010 and SPSS

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