

Cadmium (Cd) distribution and contamination in Chinese paddy soils on national scale

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Abstract Rice is a staple food by an increasing number of people in China. As more issues have arisen in China due to rice contaminated by cadmium (Cd), Cd contamination in arable soils has become a severe problem. In China, many studies have examined Cd contamination in arable soils on a national scale, but little studies have focused on the distribution of Cd in paddy fields. This study explored the spatial pattern of Cd in paddy soils in China, made a preliminary evaluation of the potential risk, and identified the most critically contaminated regions based on the domestic rough rice trade flow. The results showed that Cd concentrations in paddy soils in China ranged from 0.01 to 5.50 mg/kg, with a median value of 0.23 mg/kg. On average, the highest Cd concentrations were in Hunan (0.73 mg/kg), Guangxi (0.70 mg/kg), and Sichuan (0.46 mg/kg) provinces. Cd concentrations in paddy soils in central and western regions were higher than those in eastern regions, especially the southeastern coastal regions. Of the administrative

regions, Cd standard exceedance rate was 33.2 %, and the heavy pollution rate was 8.6 %. Regarding to Cd of paddy soil, soil environmental quality was better in Northeast China Plain than in Yangtze River Basin and southeastern coastal region. Mining activities were the main anthropogenic pollution source of Cd in Chinese paddy soil. Based on rice trade, more of the Chinese population would be exposed to Cd through intake of rice produced in Hunan province. Certain regions that output rice, especially Hunan province, should be given priority in the management and control of Cd contamination in paddy soil.

Keywords Cadmium (Cd) · Chinese paddy soil · Spatial distribution · Soil contamination · Rice trade · Risk

Introduction

China is currently facing great challenges in protecting its arable soil from contamination caused by heavy metals due to rapid industrialization and urbanization (Zhao et al. 2015). The consumption of grains, vegetables, and rhizomes planted in contaminated soil has triggered concerns about food security and human health (Liu et al. 2013). National soil contamination survey issued by the Ministry of Land and Resources Report in the 2014 shows that the main contaminants in arable soils are heavy metals, and cadmium (Cd) is the most serious contaminant in terms of exceeding the Ministry of Environmental Protection (MEP) limit (MEP 2014). As a carcinogenic contaminant, Cd has adverse effects on residents when the Cd accumulation exceeds a certain level in the body (Peralta-Videa et al. 2009). Human exposure to Cd via the food chain can cause diseases including cancers, diabetes, and high blood pressure (IARC 2004; Williams et al. 2009).

Rice is a staple food for nearly 60 % of the population of China (Shi 2015). Unfortunately, rice growing in Cd-

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contaminated paddy soils has been reported heavy Cd contamination, resulting in human health risks. In 2002, the Cd content in 10.3 % of rice products from different regions in China exceeded the limitation of environmental quality standard for soils in China (Luan et al. 2013). A survey in 2006 showed that consumption of rice growing in Cd-contaminated paddy soils threatened the health of the residents of Zhuzhou city in Hunan province (Liu 2013). Recently, in 2013, the Food and Drug Administration of Guangdong Province announced that Cd concentrations in many batches of rice and rice products were above the limitation in national standard (Luan et al. 2013).

There are large regional differences in rice producing around China (Liu et al. 2014). The Cd accumulation in paddy soils in major regions of rice production, such as Hunan and Guangdong provinces, has been studied (Wang et al. 2015; Williams et al. 2009; Zhuang et al. 2009), and the results suggest severe Cd contamination in most of China's paddy field regions. However, previous studies have largely focused on the local or regional scale (Hang et al. 2009; Wang et al. 2015; Williams et al. 2009), and understanding of the Cd contamination in paddy soils at the national scale is limited. Rice has high capacity of Cd accumulation due to its growth in paddy fields. Many studies have investigated the Cd contamination in arable land (Chen et al. 2015; Lu et al. 2015; Niu et al. 2013; Zhang et al. 2015a; Zhang et al. 2015b), but Cd contamination in flooded land such as paddy field at the national scale has not yet been investigated.

Rice trade is very actively traded markets in China due to disequilibrium of rice production (Shi 2015). Considering the domestic rough rice trade flow, we can understand the potential consumption of rice. If rice produced in Cd-contaminated area is sold to other areas, the risk distribution from Cd contamination of soil and rice may be underestimated or misjudged. Therefore, it is important to identify rice output regions in which the paddy soils have been contaminated by Cd.

In this study, we (a) determined the statistical and spatial distribution of Cd concentration in paddy soil in China, (b) evaluated contamination based on the Cd background and the national standard, and (c) conducted a contamination source analysis. Based on the results of these steps, we also identified the most critically contaminated regions according to the domestic rough rice trade flow.

Methodology

Study area

Three primary rice-producing regions, namely the Northeast China Plain, the Yangtze River Basin, and the southeastern coastal region, were identified by the Regional Planning for National Dominant Agricultural Product (2008–2015) in 2008

based on their agricultural features (MOA 2008). The Northeast China Plain region covers three Chinese provinces (Heilongjiang, Jilin, and Liaoning); the Yangtze River Basin covers ten Chinese provinces and a municipality (Sichuan, Chongqing, Yunnan, Guizhou, Hunan, Hubei, Henan, Anhui, Jiangxi, and Jiangsu), and the southeastern coastal region covers six Chinese provinces, a municipality and an autonomous region (Shanghai, Zhejiang, Fujian, Guangdong, Guangxi, and Hainan).

According to data from the China Statistics Yearbook (NBS 2015), these 19 administrative areas had the highest percentages of rice planting area in the country. The area of paddy fields in the three primary production regions (297, 288 km²) covered 98.1 % of the total paddy field area in China (303,120 km²). For the Northeast China Plain, Yangtze River Basin, and southeastern coastal region, the areas of paddy soils were 45,515, 191,619, and 60,154 km², respectively; their fractions of the total area of paddy rice in China were 15.0, 63.2, and 19.8 %, respectively.

To obtain more comprehensive results, Tianjin municipality, Shaanxi Province, and the Ningxia Hui autonomous region were also studied. In summary, a total of 22 provinces, municipalities, and autonomous regions in China were used as survey regions. In 2014, the total area of paddy soils in the 22 regions was 299,425 km², accounting for 98.8 % of the total area (303,100 km²) of Chinese paddy soils, indicating that our survey region covered most of China's paddy fields.

Data sources

Cadmium concentrations in paddy soils in China were collected from published literature on the websites of the China National Knowledge Infrastructure (CNKI) and ISI Web of Knowledge using the keywords “cadmium”, “heavy metal”, “paddy field”, “paddy soil”, and “rice” in combination with “China”. The papers were further sorted according to the following criteria: (1) the papers were published during 2000–2015; (2) topsoil samples were from layers of 0–20 cm; (3) the sampling location was clearly identified and fell within our survey region; and (4) the paddy soil was identified. Based on these criteria, 101 peer-reviewed articles were selected. Statistical databases relating to provincial paddy yields and population size were taken from the latest China Statistics Yearbook (NBS 2015), and the domestic rough rice trade flow was determined from the 2015 Report of the Chinese Rice Industry (Zheng 2015).

Data processing

Cadmium concentrations extracted from the published papers were available for 187 administrative regions of the

22 provinces, municipalities, and autonomous regions around China. The method of data processing was as follows. (1) Data extraction. We directly extracted the arithmetic average Cd concentration in each administrative region from the corresponding document. If the data in the paper were not normally distributed, the median Cd concentration was extracted. (2) Data integration. If the administrative regions in different papers were identical, we calculated the arithmetic average of the data from the papers. This arithmetic average was considered to represent the Cd content of that administrative region. If there was only one extracted data point in an administrative region, this data point was considered to represent the Cd content in that administrative region. A total of 187 Cd concentrations were obtained for the 187 administrative regions of China studied here. Details of the data are

given in Table SI-1. The spatial distributions of the samples and paddy fields in China are displayed in Fig 1. The references used as data sources are shown in the [Supplementary Information \(SI\)](#).

Pollution evaluation

Pollution index (PI)

The degree of Cd contamination was also quantitatively evaluated using a pollution index (PI), a common environmental assessment tool (Chen et al. 2015). PI was calculated using Equation (1).

$$PI = C_i/C_0, \tag{1}$$

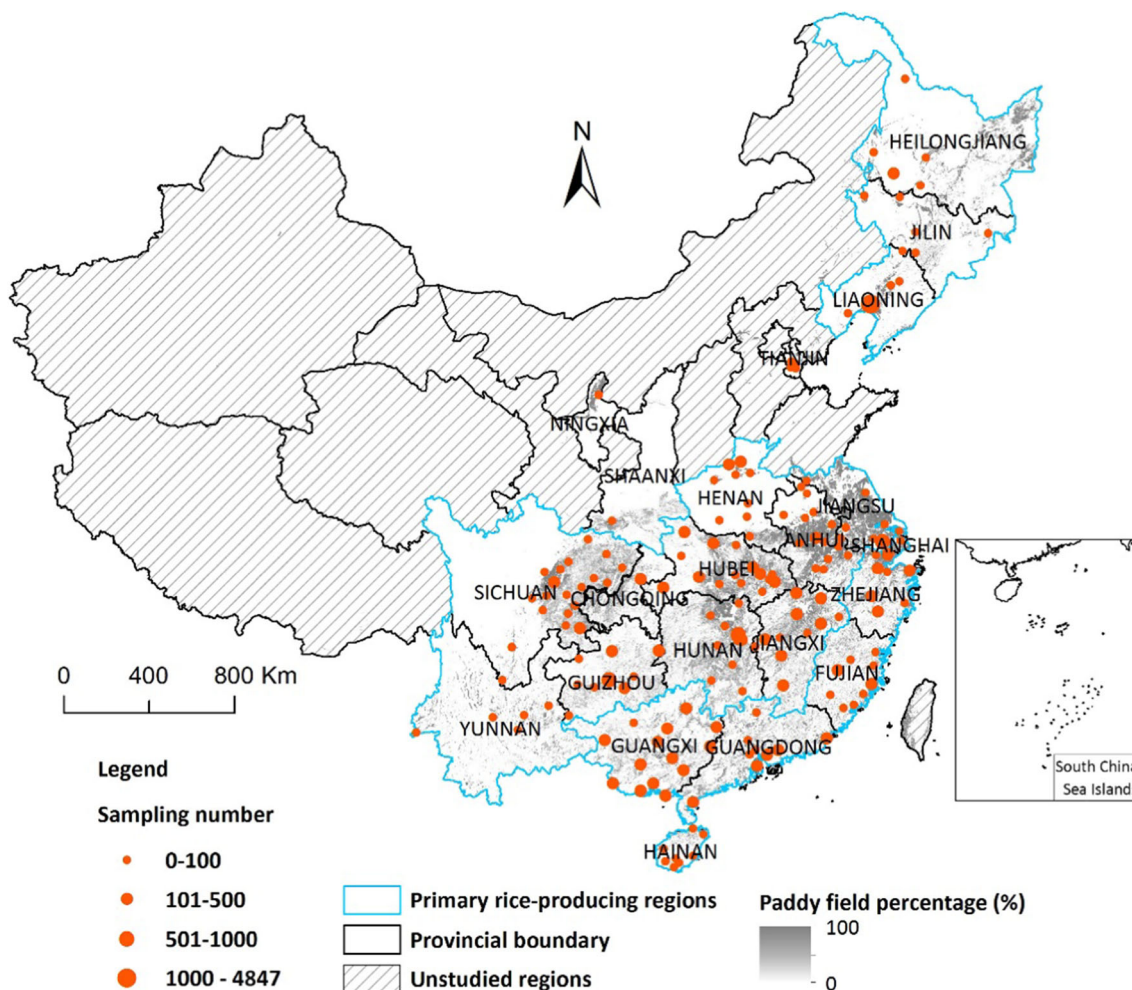


Fig 1 Spatial distribution of the sampling number and paddy field percentages in mainland China. Sampling number means the sum of sampling numbers in all data source papers. Three areas inside of the blue closed curves from north to south referred to the Northeastern China Plain region, the Yangtze River basin, and the southeastern coastal region, respectively (Hainan province belongs to the

southeastern coastal region). The gray grids represented the paddy soils distribution in China. The legend of paddy field percentage represented the percentage of rice planting area in each grid. All maps in this paper were created using geographic information system (GIS) software (ArcGIS 10.0, ESRI, Redlands, California)

where C_i is the actual concentration of Cd in a soil sample (mg/kg), and C_0 is the maximum allowable Cd level prescribed by China’s environmental quality standard for soils (mg/kg) (MEP 1995). The degree of Cd pollution based on the PI value is shown in Table 1.

Geoaccumulation index (I_{geo})

The geoaccumulation index (I_{geo}), originally introduced by Müller (1969), can be used to evaluate the environmental pollution status compared with preindustrial concentrations. Recently, it has been widely applied in contamination assessments of heavy metals in soil, dust, water, and sediment (Bhuiyan et al. 2010; Lu et al. 2009; Suthar et al. 2009). In this study, the geoaccumulation index was calculated using Equation (2):

$$I_{geo} = \log_2 \frac{C_i}{1.5B_n} \tag{2}$$

where C_i is the Cd concentration in the soil samples (mg/kg) and B_n is the national geochemical background value of Cd in the soil (mg/kg) (CNEMC 1990; Geng et al. 2012; Ji 2006; Zhao 2008). The geoaccumulation index consists of seven grades, which represent seven degrees of pollution. The relationship between the I_{geo} value and the degree of pollution is shown as Table 2.

I_{geo} -PI

We divided the pollution level into four types based on the I_{geo} value combined with the PI. The detailed method is shown in Table 3.

Results

Cadmium distribution in the paddy soils of China

The total paddy field area in China was 303,100 km², accounting for 18.3 % of the national arable area (1,654,460 km²) in 2014 (NBS 2015). The Cd concentrations in paddy soils ranged from 0.01 to 5.50 mg/kg, with a median of 0.23 mg/kg, standard deviation (SD) of 0.76 mg/kg, and coefficient of variation

Table 1 Percentages of class distribution for pollution assessment of Cd in Chinese paddy soils using PI value ($n = 187$)

PI value	Pollution degree ^a	The number of administration region and its percentage
PI ≤ 1	Clean	125, 66.8 %
1 < PI ≤ 2	Light pollution	22, 11.8 %
2 < PI ≤ 3	Moderate pollution	24, 12.8 %
PI > 3	Heavy pollution	16, 8.6 %

^a Pollution degree refers to Chen et al. 2015 and Song et al. 2013

Table 2 Percentages of class distribution for pollution assessment of Cd in Chinese paddy soils using I_{geo} value ($n = 187$)

I_{geo}	Pollution degree	The number of administration region and its percentage
$I_{geo} \leq 0$	unpolluted	54, 29.0 %
$0 < I_{geo} \leq 1$	unpolluted to moderately polluted	76, 40.6 %
$1 < I_{geo} \leq 2$	moderately polluted	38, 20.3 %
$2 < I_{geo} \leq 3$	moderately to heavily polluted	7, 3.7 %
$3 < I_{geo} \leq 4$	heavily polluted	7, 3.7 %
$4 < I_{geo} \leq 5$	heavily to extremely polluted	3, 1.6 %
$I_{geo} > 5$	extremely polluted	2, 1.1 %

(CV) of 1.69 ($n = 187$) (Table 4). Because the dataset did not follow a normal distribution (Fig SI-1), the median was preferable to the arithmetic mean value for evaluating the average content (Guo 2006). The large range of the data and the high SD and CV indicated that Cd concentrations in the paddy soils varied considerably among the different regions. Average Cd in Chinese arable land has reported by some studies including 0.27 mg/kg from Zhang et al. (2015a) and 0.21 mg/kg from Niu et al. (2013). Thus, the average Cd level in Chinese paddy soil was similar to that in Chinese arable soil.

On average (Table 4), the highest Cd concentrations were in Hunan province (0.73 mg/kg) in central China, Guangxi province (0.70 mg/kg) in the southwest, and Sichuan province (0.46 mg/kg) in the west. The lowest Cd concentrations were in Henan (0.02 mg/kg) in northern China, Heilongjiang (0.09 mg/kg) in the northeast, and Hainan (0.09 mg/kg) in the south. The spatial distribution of Cd concentrations in paddy soils across China indicates that Cd levels varied widely among the regions (Fig 2). In general, Cd concentrations in paddy soils in the central and western regions were higher than those in the eastern regions, especially the southeastern coastal regions.

Cd levels in the three dominant rice-producing regions

According to the Regional Planning for National Dominant Agricultural Product (2008–2015) (MOA, 2008), the division

Table 3 Percentages of class distribution for pollution assessment of Cd in Chinese paddy soils using PI and I_{geo} ($n = 187$)

PI, I_{geo}	Pollution evaluation	The number of administration region and its percentage
PI < 1, $I_{geo} \leq 0$	Non-pollution and no Cd introduced from human activities	52, 27.8 %
PI < 1, $I_{geo} > 0$	Non-pollution but Cd has been introduced from human activities	72, 38.5 %
PI > 1, $I_{geo} \leq 0$	Pollution caused by soil background	1, 0.5 %
PI > 1, $I_{geo} > 0$	Pollution caused by human activities	62, 33.2 %

Table 4 Descriptive statistics for Cd concentration in paddy soils

Province/Municipality/ Autonomous region	Sample number ^a	Mean (mg/kg)	SD ^b	CV ^c	Min (mg/kg)	Q1 ^d (mg/kg)	Median (mg/kg)	Q3 ^e (mg/kg)	Max (mg/kg)	PI ^f	<i>I</i> _{geo}
Nationwide	187	0.45	0.76	1.69	0.01	0.14	0.23	0.46	5.50	0.77	0.66
Heilongjiang	5	0.09	0.02	0.25	0.06	0.08	0.09	0.11	0.11	0.30	-0.52
Jilin	9	0.22	0.14	0.65	0.11	0.19	0.19	0.19	0.60	0.63	0.36
Liaoning	5	0.83	1.06	1.27	0.14	0.14	0.19	1.12	2.57	0.63	0.23
Sichuan	22	0.86	1.04	1.21	0.06	0.35	0.46	0.95	4.76	1.53	1.96
Chongqing	1	0.39	—	—	0.39	0.39	0.39	0.39	0.39	1.30	0.97
Yunnan	5	0.45	0.47	1.04	0.21	0.25	0.25	0.25	1.28	0.83	-0.39
Guizhou	9	0.32	0.12	0.38	0.22	0.25	0.27	0.34	0.61	0.90	-1.87
Hunan	10	1.12	1.45	1.30	0.45	0.47	0.73	0.75	5.21	2.43	1.95
Hubei	15	0.28	0.43	1.54	0.09	0.11	0.12	0.18	1.74	0.40	-1.10
Henan	8	0.06	0.09	1.40	0.02	0.02	0.02	0.06	0.27	0.03	-2.47
Anhui	15	0.46	1.08	2.33	0.10	0.13	0.17	0.27	4.36	0.57	0.22
Jiangxi	11	0.25	0.18	0.71	0.01	0.11	0.18	0.41	0.61	0.60	0.15
Jiangsu	13	0.25	0.07	0.26	0.12	0.26	0.26	0.26	0.41	0.43	0.46
Shanghai	2	0.23	0.13	0.55	0.14	0.14	0.23	0.32	0.32	0.77	0.15
Zhejiang	11	0.22	0.04	0.18	0.17	0.20	0.21	0.21	0.31	0.70	1.00
Fujian	9	0.17	0.03	0.17	0.14	0.15	0.16	0.20	0.22	0.53	0.53
Guangdong	9	0.92	1.74	1.90	0.05	0.20	0.20	0.81	5.50	0.67	1.25
Guangxi	14	0.74	0.13	0.18	0.70	0.70	0.70	0.70	1.20	2.33	0.81
Hainan	8	0.08	0.03	0.34	0.03	0.07	0.09	0.11	0.11	0.30	1.15
Tianjin	4	0.25	0.08	0.33	0.17	0.20	0.23	0.30	0.36	0.28	0.33
Ningxia	1	0.19	—	—	0.19	0.19	0.19	0.19	0.19	0.32	0.18
Shaanxi	1	0.17	—	—	0.17	0.17	0.17	0.17	0.17	0.38	0.71

^a Sample number equals to the number of administrative regions;
^b Standard deviation;
^c Coefficient of variation;
^d 1st quartile, 25 %;
^e 3rd quartile, 75 %;
^f PI and *I*_{geo} was calculated with the Cd median value of paddy soils in each province

of primary rice-producing regions is based on regional differences in rice variety and rice yield. The median concentration of Cd in paddy soil in the Northeast China Plain, Yangtze River Basin, and southeastern coastal region was 0.19, 0.26, and 0.21 mg/kg, respectively, (Table 5). The average Cd content in the Yangtze River Basin was higher than that in China overall (0.23 mg/kg). The average Cd content in the other two regions was lower than that in China. High CV values for the Cd concentration in these three regions (1.70, 1.68, and 1.71, respectively) demonstrated that the Cd concentration in each rice-producing region varied greatly.

Cadmium accumulation in rice is dependent on the rice variety (Zhao et al. 2009), and the consumption of the rice produced is driven by rice yield (Liu et al. 2014; Shi 2015). The domestic rice trade has a close relation with the rice-producing regions (Zheng 2015). For example, three provinces in northeastern China are considered as one large region in terms of rice output (Zheng 2015). So, it is significant to analyze the Cd distribution in the three dominant rice-producing regions of China.

Pollution evaluation based on the PI and *I*_{geo}

The PI values for Cd ranged from 0.03 to 18.3, with a nationwide median value of 0.67 (*n* = 187, mean = 1.46, SD = 2.58,

CV = 1.76). Of the PI values, 33.2 % exceeded 1 (62 of 187 administrative region-scale samples), meaning that the Cd concentrations in paddy soils in 33.2 % of all administrative regions exceeded the Chinese Cd threshold (0.3 mg/kg when pH ≤ 7.5; 0.6 mg/kg when pH > 7.5). Zhang et al. (2015a) reported that the Cd concentrations in 26.3 % of samples exceeded the maximum permissible concentrations of 0.60 mg/kg. Song et al. (2013) investigated Cd pollution in the farmland of 138 administrative regions, showing that Cd in 25.2 % of all regions exceed the Chinese soil II standard. However, no published data were available for Cd concentrations in paddy soil at a national scale. This indicates that the average Cd contamination was higher in Chinese paddy soil than in Chinese arable soil. In the three primary rice-producing regions of the Northeast China Plain, Yangtze River Basin, and the southeastern coastal region, the percentage of Cd measurements that exceeded the standard value was 15.8 % (3 of 19), 36.7 % (40 of 109), and 35.8 % (19 of 53), respectively. Cd contamination of paddy soil in the Yangtze River Basin and southeastern coastal region was more serious than the Northeast China Plain. On average of each province (Table 4), PI values of Hunan (2.43), Guangxi (2.33), Sichuan (1.53) provinces, and Chongqing municipality (1.30) exceeded 1. That is on average, the paddy soils in Hunan, Guangxi, Sichuan provinces, and Chongqing municipality

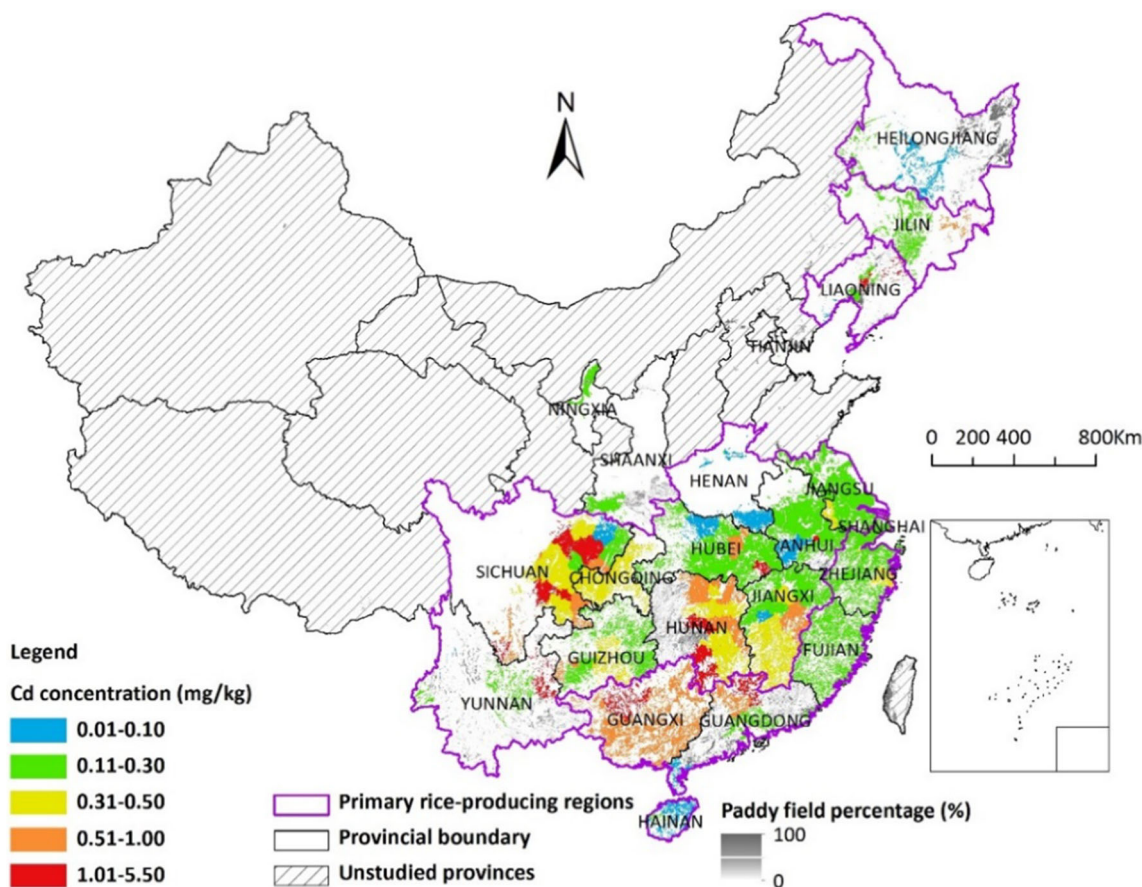


Fig 2 Spatial distribution of Cd concentration in Chinese paddy soils at the administrative region scale. Three areas inside of the violet closed curves from north to south represented the Northeastern China Plain region, the Yangtze River basin, and the southeastern coastal region,

respectively, (Hainan province belongs to the southeastern coastal region). No Cd concentration in the blank areas was attributed to no paddy soil distribution

were contaminated by Cd compared to national soil environmental quality standard.

According to the National Soil Contamination Survey results from the Ministry of Land and Resources Report in 2014, 19.4 % of samples exceeded the Chinese standard for agricultural soils, and Cd was the most severe contaminant. Among all samples, 7 % exceeded the standard for Cd. This was the highest exceedance rate of all heavy metals. According to survey results, it was apparent that the Cd pollution was more serious in the south than in the north. Pollution problems in parts of the Yangtze River Delta, Pearl River Delta, and the

northeastern industrial region were more severe. Areas of pollution in the southwest and central-south regions were more extensive. Our study (Fig 1) indicated that the primary rice-producing regions in China were distributed in arable land with more serious contamination, as indicated by the 2014 National Soil Contamination Survey from the Ministry of Land and Resources Report.

According to the degree of Cd pollution based on the PI value (Table 1), the paddy soils in 66.8 % of regions were in the clean category, 11.8 % showed light pollution, 12.8 % had moderate pollution, and 8.6 % had heavy pollution. From Fig 3,

Table 5 Descriptive statistics for Cd concentration in paddy soils of primary rice-producing regions

Rice-producing regions ^a	Sample number ^b	Mean	SD	CV	Min	Q1	Median	Q3	Max
NP	19	0.35	0.59	1.70	0.06	0.11	0.19	0.19	2.57
YRB	109	0.49	0.82	1.68	0.01	0.14	0.26	0.45	5.21
SC	53	0.45	0.76	1.71	0.03	0.16	0.21	0.70	5.50

^a There are three primary rice-producing regions. NCP, YRB, and SC represent the region of Northeastern Plain, Yangtze River Basin, and southeastern coastal region, respectively

^b Sample number equals to the number of administrative regions

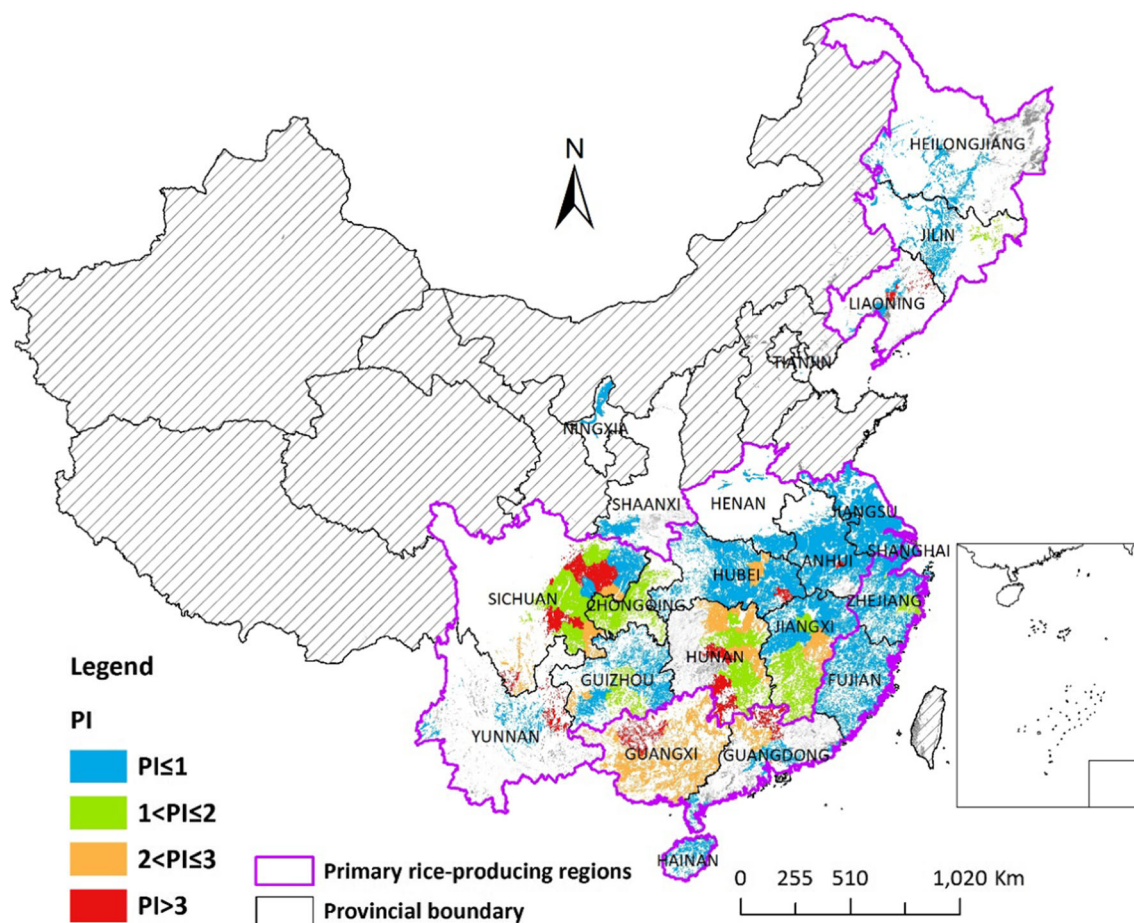


Fig 3 Spatial distribution of Cd pollution in Chinese paddy soil based on the PI values

it can be seen that paddy soils considered clean with respect to Cd were mainly located in the northeastern and eastern regions, especially the coastal regions. Lightly polluted paddy soils for Cd were mainly located in the western and central regions. Moderately polluted paddy soils for Cd were mainly located in the southern and central regions. Heavily polluted paddy soils for Cd were dispersed over a wide area, including Liaoning, Anhui, Hunan, Hubei, Guangdong, Guangxi, Yunnan, and Sichuan Provinces.

The nationwide I_{geo} values for soil Cd ranged from -4.02 to 6.03 ($n = 187$), with high SD (1.42) and CV (2.13) values. Of the I_{geo} values, 71 % exceeded 0 (133 of 187 administrative region-scale samples), meaning that Cd had been introduced from human activities to paddy soils in 71 % of all administrative regions. Table 2 gives the percentage of contaminated regions based on I_{geo} values.

Using the average Cd concentration (0.23 mg/kg) in paddy soil of China, we calculated the I_{geo} and PI values in China to be 0.661 and 0.767, respectively. This result demonstrated that Cd had been introduced into paddy soils from human activities, but the average pollution level was still within the acceptable range for the environment in China. A pollution evaluation based on I_{geo} -PI values is shown in Fig SI-2. Only in one

region of central Guizhou was the high level of Cd due to the geogenic soil, not to human activities. The sample number of “PI < 1, I_{geo} > 0” was 72 (Table 3), which accounted for 58 % of clean soil samples and 38.5 % of total samples. “PI < 1, I_{geo} > 0” indicated that the paddy soils in these regions had not been polluted, but Cd had been introduced from human activities. These regions were mainly distributed in the southeastern coastal area. In summary, Cd pollution in paddy soils in China was mainly due to human activities.

Anthropogenic pollution sources analysis

Figure 4 shows the distribution of Cd pollution source in paddy soil according to our data. Pollution sources were specific in 28 administrative regions, i.e., 15 % of the 187 administrative regions. The pollution sources included the mining and smelting industry, major industrial areas, sewage irrigation, and electronic waste disposal. Cd contamination in 16 administrative regions was due to mining. This number accounted for 57 % of the 28 regions with specific pollution sources. Mining activity was the most important pollution source, with mining areas located in Anhui, Hubei, Jiangxi, Hunan, Guangdong, Guangxi, and Yunnan. Moreover, 56 % of

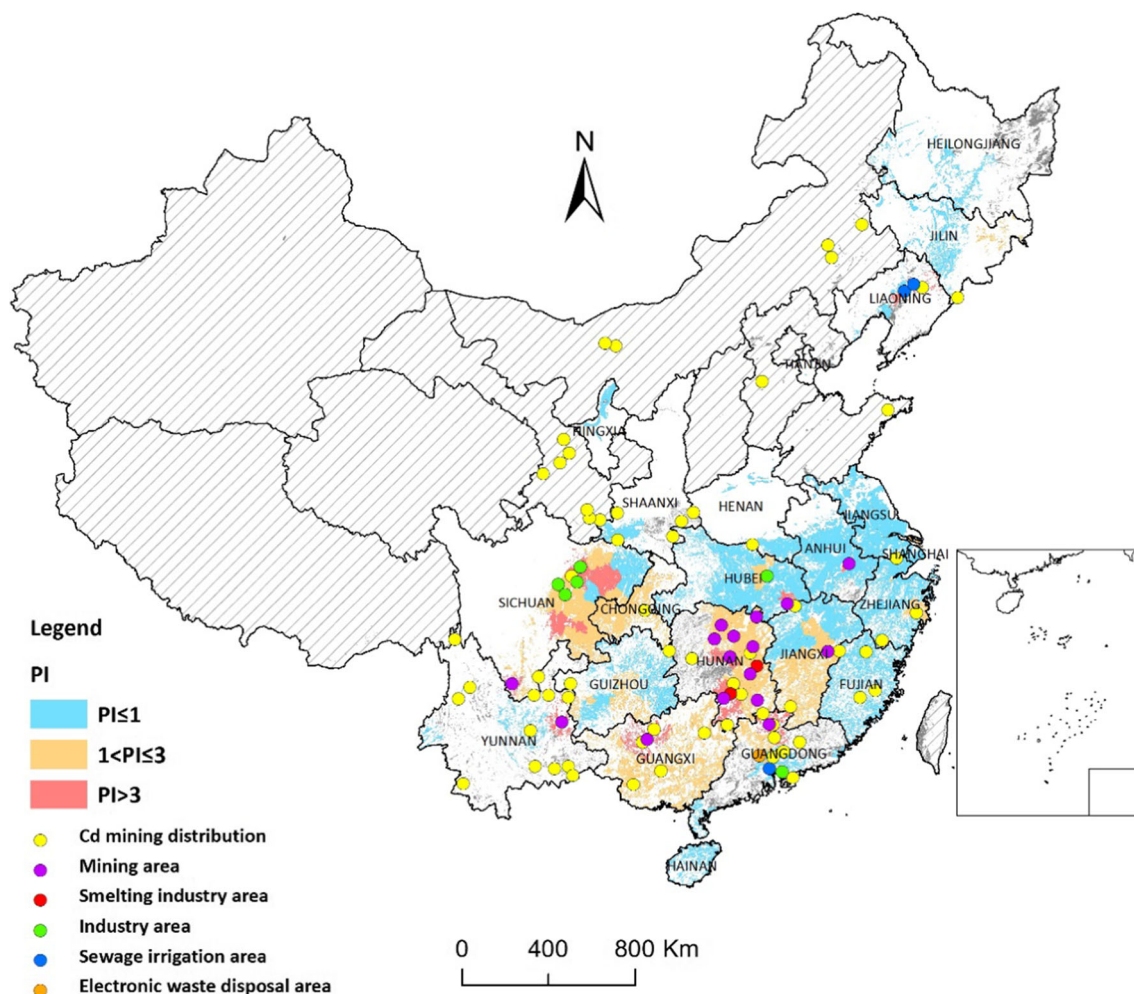


Fig 4 Source distributions of Cd pollution in paddy soil. The *yellow dot* referred to the distribution of Cd mining area (Cd deposits higher than 500 ton) in China (Yuan et al., 2012). Other *color dots* represented Cd pollution sources in this study

mining areas were located in Hunan province. Industrial areas were distributed in Sichuan, Hubei, and Guangdong. Sewage irrigation areas were distributed in Liaoning and Guangdong. Cd contamination in Liaoning was mainly sourced from sewage irrigation in the old industrial areas of the northeast. Qingyuan city in Guangdong province is a well-established electronic waste disposal area, and paddy soils have been contaminated by this source. From Fig 4, it can be seen that greater aggregation of pollution sources is associated with higher PI values.

Zhang et al. (2015a) discussed three sources of Cd contamination in Chinese arable land: mining and smelting activities, wastewater irrigation, and urban activities. Their study demonstrated that soil contaminated by the mining and smelting industries had higher Cd concentrations than soil contaminated by other sources. Our research also showed that mining activity was the main pollution source for Cd contamination in Chinese paddy soils (57 % of all contaminated soils were from mining areas). Many other studies have also reported that severe Cd contamination in croplands in

China is due to mining activities (Lu et al. 2015; Zhao et al. 2015). In fact, much of the Cd contamination was due to non-standard mining methods and smelting processes (Lu et al. 2015), such as mineral excavation, ore transportation, mineral smelting, and wastewater disposal (Zhang et al. 2015a). Agricultural systems, including arable land and irrigation water, would be affected by environmental pollution from mining activities (Lu et al. 2015). Figure 4 shows the Cd-bearing mining areas in China according to Yuan et al. (2012). In China, Cd minerals are mainly distributed in the central, southwestern, and eastern regions, and the total Cd deposits in these areas account for 88 % of the proven national deposits (Yuan et al. 2012). Fujian province in the southeastern coastal region has abundant Cd deposits, but our investigation indicated that the average Cd concentration in Fujian was 0.16 mg/kg (Table 4), which is lower than the standard. More frequent pollution events have occurred in Hunan, Guangxi, and Guangdong provinces, and Cd contamination is more serious in these regions (Table 4).

Determination of risk and the most critically polluted regions of China based on the rough rice trade flow

Rice is the staple food for an increasing number of people in China, and the rice yield has increased year by year since 2002 (Shi 2015). In 2014, China’s rough rice yield was 206.51 million tons (NBS 2015), and the output and input quantities were 0.63 tons and 3.90 million tons, respectively (Zheng 2015). There are large regional differences in rice production in China (Fig SI-3). The rough rice yield in nine regions exceeded 10 million tons in 2014. According to the Report of Chinese Rice Industry in 2015 (Zheng 2015), three provinces in the northeast (Heilongjiang, Jilin, and Liaoning provinces), four provinces in the central-south (Anhui, Hubei, Hunan, and Jiangxi provinces) and Jiangsu province had rough rice surpluses to output, and Zhejiang, Fujian, Guangdong, and Guangxi provinces in the coastal region had large deficits (Fig 5). A rough rice trade flow has formed in China over recent decades (Zheng, 2015).

Among eight major regions of rice output (Fig 5), the average Cd level of paddy soil in Hunan was highest (0.73 mg/kg). In Hunan, the PI values of the regions impacted by mining activities were much higher than 1 (2.43). The average Cd levels of paddy soils in other output regions were lower than the standard (PI < 1) (Table 4). Potential human risk across China would be introduced from Hunan province through rice trade. Hunan province had the highest yield among all the provinces (26.24 million tons in 2014) (Fig SI-3). Rough rice in Hunan is exported to the western and southeastern coastal regions. The population density in Guangdong and Zhejiang exceeds 500 persons/km². So more Chinese population are exposed to Cd through the dietary intake of rice produced in Hunan province. There are indications that the regions and population outside of contaminated regions have been affected by the Cd contamination of paddy soils. In terms of control measures, priority should be given to managing the regions of rice output, especially Hunan province.

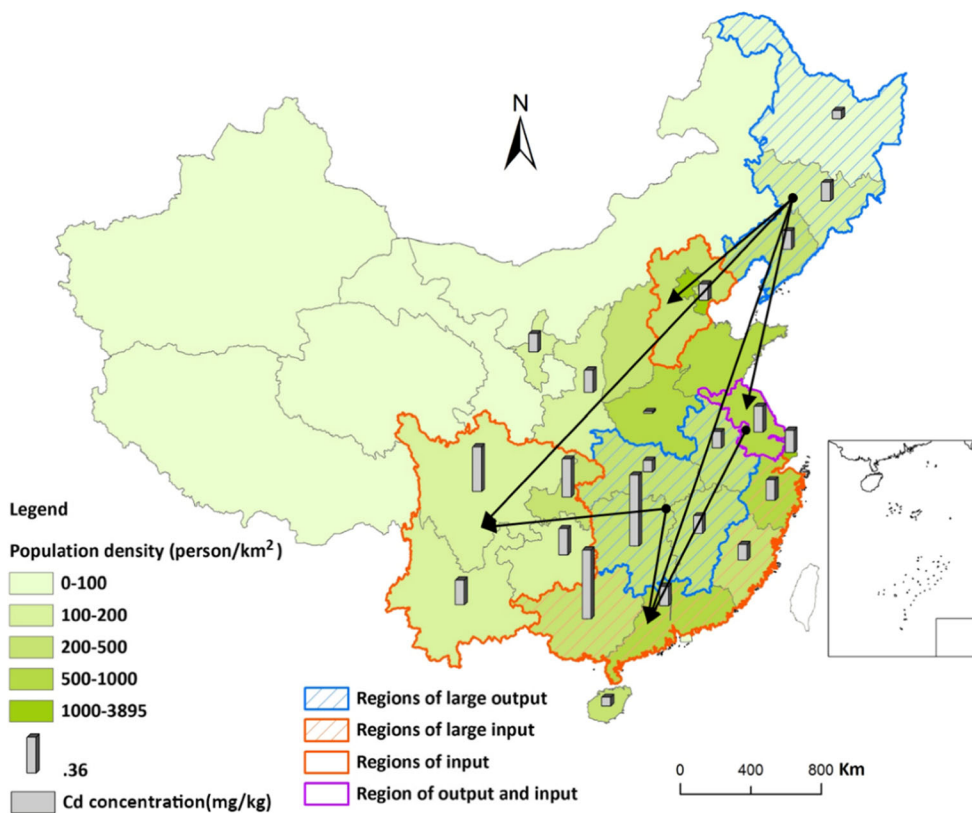


Fig 5 Trade flow of rough rice in mainland China. Cd concentration was the median value of administrative regions in each province. Regions of large output which had the largest rice surplus in China included two large regions: the northeastern large region and the central-southern large region. The northeastern large region was the northeastern plain region including Heilongjiang, Jilin and Liaoning provinces, and the central-southern large region covered four provinces which were Jiangxi, Hunan, Hubei, and Anhui provinces. Regions of large input which had

largest rice gaps in China covered four provinces: Zhejiang, Fujian, Guangdong, and Guangxi. The *black arrows* meant the rough rice output flows. The data of rough rice yield was source from China statistics yearbook (NBS, 2015). Rough rice trade flows referred to the Report of Chinese Rice Industry in 2015 (Zheng, 2015). This map was created using Geographic Information Systems software (Arc GIS version 10.2, ESRI Inc., Redlands, Calif)

In addition, China's rough rice imported from other countries accounted for 1.9 % of the national total yield in 2014, 99 % of which was from Thailand (NBS 2015; Shi 2015). The International Water Management Institute (IWMI) investigated 524 paddy soils in Thailand and found that Cd concentrations were 0.5–284 mg/kg (Simmons et al. 2005). Kosolsaksakul et al. (2014) stated that paddy fields were severely contaminated by zinc ore extraction in the Mae Tao watershed of northwest Thailand, and Cd contents there ranged from 2.5–87.6 mg/kg. Nevertheless, Cd background concentrations in Thai soils were only 0.002–0.14 mg/kg (Pongsakul and Attajarusit 1999). The safety of foreign rice products and their paddy soils should not be ignored.

Limitations of this study

This study had three limitations. First, our data did not cover all areas of Chinese paddy soil due to the lack of available Cd concentration data in some areas (Fig 1). Second, there were differences in the sampling and analysis strategies of the references consulted. Third, some references specifically targeted potentially contaminated sites, adding a source of bias to the dataset.

Discussion

In this study, we analyzed the Cd contamination in paddy soils of China at national scale. The average Cd concentration in paddy soils was 0.23 mg/kg, and the CV was high (1.69) (Table 4). The Cd standard exceedance rate was 33.2 %, and the heavy pollution rate was 8.6 %. The contaminated regions were distributed throughout the country (Fig 3). Although the average Cd level in Chinese paddy soil was similar to that in Chinese arable soil in general, the Cd standard exceedance rate was higher. This further indicated that Cd concentrations in paddy soils from different Chinese regions varied significantly. All of the Cd pollution was from human activity, except in one administrative region in Guizhou, where the geogenic soil Cd was the main factor. The mining industry was the major anthropogenic pollution source, with pollution incidents from mining activities being the direct cause and critical driving force of Cd contamination. From Table 4, the highest Cd levels appeared in Hunan (0.73 mg/kg, PI = 2.43) and Guangxi (0.70 mg/kg, PI = 2.33) provinces were mainly induced from mining activities (Chen et al. 2015; Williams et al. 2009). Besides that, Cd average levels of paddy soils in Sichuan province and Chongqing municipality were also high (0.46 and 0.39 mg/kg). Li (2014) discussed the impacts of social and economic development on Cd of farmland in Chengdu city of Sichuan province, including the increased industrial enterprises, excessive fertilizer application, and the

development of transportation industry. However, investigation on Cd contamination source of arable soil in Sichuan province had not carried out and contamination source had not been identified (Jin et al. 2012).

Soil characteristics were the important factors for the distributions of heavy metals in soil (Lu et al. 2015; Zhao et al. 2015). Soil pH is considered an important factor affecting the migration and transformation of heavy metals (Bradl 2004). Soil pH is one of the most important factors causing high levels of Cd accumulation in rice (Kögel-Knabner et al. 2010). Takijima et al. (1973) showed that Cd solubility was minimal at a soil pH of 7. Zhao et al. (2015) indicated that the Cd content of rice grown on non-polluted soils in many areas of southern China would exceed the Cd standard due to the extreme acidity of paddy soils. The spatial patterns of soil pH in China are presented in Fig SI-4, which shows significant differences across areas in China. Overall, the areas of paddy soil in China are covered by acid soils. Further research is required to determine the potential relationships between soil pH and soil type. Moreover, Guo et al. (2010) indicated that the average pH declined by 0.13 ($P < 0.001$) in paddy soils in cereal crop systems between the 1980s and 2000s. Hunan province currently has the largest area of soil acidification in China. Two-thirds of the farmland in that province has experienced various degrees of soil acidification (Mo 2013). Significant soil acidification on cropland topsoil would result in severe issues of rice safety in Hunan, where Cd levels in paddy soil are high.

Cadmium contamination in paddy soil has an influence on rice yield and quality, causing crop yield to decrease and grain quality to decline (Lu et al. 2015). This is clearly detrimental for rice production and the national economy. Therefore, in the face of the serious situation regarding paddy soil contamination, contamination control measures should be adopted in China. Recently, China's central leaders made a preliminary decision to incorporate the environmental quality of national farmland into the 13th Five Year Plan (CPC CC 2015). However, the cost of soil remediation is known to be very high. Unlike in urban soil remediation, the government and farmers cannot endure the high cost of cropland soil remediation because the land use (i.e., crops) cannot provide enough value to subsidize the remediation. Therefore, we should identify significant regions according to regional Cd contamination levels and consider the scope of the impact of Cd contamination. In our study, Hunan province had especially significant influences on national rice safety. If necessary, isolation or the prohibition of cultivation in contaminated regions can also be recommended (Zeng et al. 2013). However, the lack of a quantitative evaluation of the potential risk and of the most critically contaminated regions was a significant limitation of our study. This should be the aim of future research.

Conclusion

In conclusion, we analyzed the Cd distribution in Chinese paddy soils on a national scale, and the Cd levels in paddy soils of the three primary rice-producing regions. The spatial distribution showed that Cd concentrations in paddy soils in the central and western regions were higher than those in the eastern regions, especially the southeastern coastal regions. We evaluated Cd contamination levels in paddy soils. Of the administrative regions, Cd standard exceedance rate was 33.2 %, and the heavy pollution rate was 8.6 %. Cd contamination was more severe in paddy soils than in other types of arable soil. Regarding to the average level of each province, the paddy soils in Hunan, Guangxi, Sichuan provinces, and Chongqing municipality were contaminated by Cd compared to national soil environmental quality standard. Soil environmental quality is better in Northeast China Plain than in Yangtze River Basin and southeastern coastal region for Cd in paddy soil. Since Cd contamination of paddy soil was mainly from human activities, the impact of mining and other human activities on Cd concentration was analyzed. Results indicated that mining was the main source of Cd contamination in Chinese paddy soils, and pollution incidents from mining activities were the critical driving force. Finally, we identified critically contaminated regions of Hunan province based on the rough rice trade flow. Cadmium contamination in paddy soils in Hunan province should be given priority in terms of management and control.

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