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Distribution Characteristics of Heavy Metals in Different Particle Size Fractions of Chinese Paddy Soil Aggregates

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Abstract: In recent years, the migration and transformation of heavy metals (HMs) in soil has become a hot issue. Soil particle size has an important effect on the environmental behavior of HMs in soil. The distribution of HMs in soil is strongly affected by the size of a soil aggregate. In this study, paddy samples in both cultivated and uncultivated soils were collected from Anhui Province, China. The soils were sieved into six particle size fractions (diameters of >4000, 4000–2000, 2000–1000, 1000–250, 250–53, and <53 μm) and the wet digestion method was used to determine the concentrations of Cd, Cr, Pb, and As in the soil aggregates. The results showed that the surface soils were characterized by the largest proportion of coarsest aggregates of >4000 μm particles. Moreover, the concentrations of Cd, Cr, Pb, and As were elevated in cultivated paddy soils compared to uncultivated soils. The accumulation of HMs in all paddy soils increased with decreasing particle size. Although the smallest particle size fractions accounted for only 5.65–17.28%, they provided the highest distribution factor (DF) of Cr (1.35), As (1.25), Cd (1.28), and Pb (1.38). The highest contributions of HMs came from the coarser fractions (>2000 μm); however, for Cr, Pb, Cd, and As, the mass loading of <53 μm particles was up to 15.90%, 14.41%, 15.21%, and 15.70%, respectively. The highest content of HMs was found in the finest aggregate, with a pattern of decreasing with increasing aggregate size. In addition, the leachability behaviors of different HMs in different particle size aggregates were different. The leachability behaviors of Cr and Pb in the coarse particle size fractions were the highest, while the leachability behaviors of As and Cd in the less than 250 μm particle size fractions were 10–100 times higher than those of Cr and Pb. A correlation analysis showed that the particle distributions of Fe, TOC, and Mn had significant influences on the distributions of Pb, Cd, As, and Cr. This study provides a theoretical basis for the prevention, treatment, and remediation of HMs pollution in soil.

Keywords: particle size fraction; paddy soil; heavy metals; soil aggregates; leachability

1. Introduction

With the rapid development of industry and agriculture as well as increasing population, high-intensity economic activities have had a significant impact on the farmland environment. Heavy metal (HM) pollution of farmlands is becoming a hot issue of global concern [1–4]. In recent years, significant attention has been focused on heavy metal pollution in paddy soil. A national survey carried out by the Chinese government, in 2014, presented that the concentrations of Cd, Hg, As, Cu, Pb, Cr, Zn, and Ni in soil were quite high, exceeding rates of 1.6%, 2.7%, 2.1%, 1.5%, 1.1%, 0.9%, and 4.8%, respectively [5]. Paddy soil is an important agricultural soil, covering nearly 140 million hectares of the Earth's surface and providing food for more than 50% of the total population on the Earth [6]. In recent years, HM pollution in rice fields has been widely reported [7–9]. Zinc mining activities in northern Thailand have led to severe Cd pollution in paddy soils [10]. In Hunan Province, one of the most important rice planting areas in China, it

has been reported that the contents of Cd, As, and Pb in rice samples from paddy fields affected by mines exceeded the Chinese national food standards by 65%, 50%, and 34%, respectively [6,11].

A soil aggregate is the elementary unit of soil systems [12]. The content of HMs in a soil aggregate has a strong correlation with particle size [13–15], which in turn affects the mobility and biological effectiveness of soil HMs. According to the Chinese domestic and foreign classification standards, soil aggregates can be divided into fine aggregates with a particle size of <250 μm and coarse aggregates with a particle size of >250 μm [12]. Fine aggregates present a high specific area and more HMs can be adsorbed [6,16–18]. Moreover, fine soil fractions are often preferentially transported to surface water and deep soil [16,19,20]. Under the influence of external factors (e.g., rain, fertilizer, etc.), HMs in fine soil fractions can easily accumulate in deep soil, groundwater, or surface water. Many studies of HMs in particle size fractions have been reported in urban soil [21–23], garden soil [24], dust [21,25,26], and sediment [27–29] and the results have indicated that the concentration of HMs in soils increased with decreasing particle size [30,31]. The results have also shown that paddy soil aggregates with clay-sized particles have the largest adsorption capacity for Cu and Zn because they contain higher organic matter, iron, and manganese oxides [32]. However, other studies have found high levels of Cu, Zn, Cd, and Pb in coarse aggregates of rice field soil particles in southern China that have long been contaminated with HMs [33]. It has been shown that in polluted paddy soil, the distribution of HMs in different particle size aggregates is complex [6].

In paddy soil, periodic artificial drainage and ploughing can lead to significant changes in the physical and chemical conditions of the vertical profile [6]. Due to the destruction of surface structure and changes in redox conditions, clay particles are enhanced to migrate to the bottom of the plough layer, which helps to form a plough disk under additional mechanical pressure exerted by buffalo or tractors [34]. In most cases, exogenous metals are first retained in the surface soil, and then transported to the deep layer through the leaching process, resulting in a gradual decrease in the metal concentration with an increase in soil depth. In addition, some researchers have also pointed out that the spatial distribution of HMs is closely related to the clay content in paddy soils [6]. The contents of total organic matter and HM_S in soil have been shown to vary with the size of soil particles, preferring to exist in silty- and clay-soil particles, which leads to significant variations of HM leachability in different particle soils [35].

To date, there have been relatively few studies on the distribution of HMs in paddy soil particles, especially the fluidity of HMs in different particle size aggregates. In this study, the pollution index was used to evaluate the pollution degree of Cr, Pb, As, and Cd in paddy soil with different grain sizes. The distribution and toxic leaching of HMs with different particle sizes in arable soil were studied by using distribution coefficients and HM mass loading. The leachability of HMs with different particle sizes in arable soil was studied by using high purity water (pH = 7). The information provided in this paper is of significance for further understanding the environmental behavior of HMs in paddy soil and formulating reasonable remediation strategies to control related risks.

2. Material and Methods

2.1. Study Area and Sampling

Anhui Province (29°41′–34°38′ N, 114°54′–119°37′ E) is located in the southeast of China, situated between the Huaihe River and Yangtze River. The central area has a warm and humid subtropical climate with a 14–17 °C mean annual temperature range and 1200 mm of precipitation. These conditions make the area an important production base for grains, and more than 70% of the cultivated soil is used for paddy rice farming.

The sampling sites for this study are shown in Figure 1. The surface paddy soils (0–20 cm) were collected from three areas (Shouxian, Feidong, and Shucheng) located in the center of Anhui Province, and the samples were collected from cultivated sites. Five subsamples were collected from a 20–50 m region around each sampling point and mixed

into one sample. The samples were kept in plastic boxes. Reference samples were collected from wasteland covered with weeds and shrubs and without anthropogenic activities near the sampling site for soil pollution assessment. All samples were air dried at room temperature and coarse debris was removed. All samples were stored in a refrigerator at 4 °C until further processing. The remaining water content in the soil was determined gravimetrically after drying individual subsamples at 105 °C overnight. All the results were reported as dried weight.
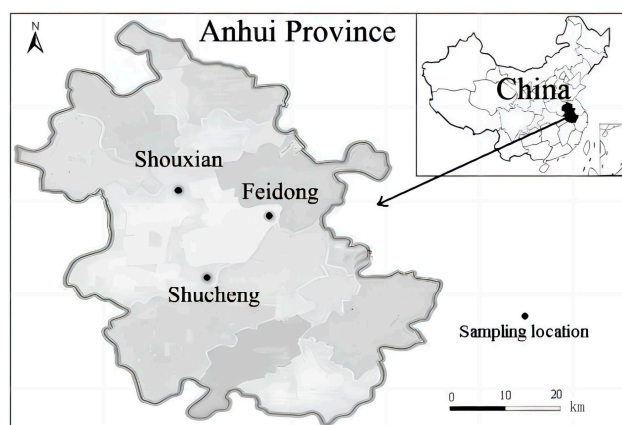

Figure 1. Soil sampling location in the study area.

2.2. Analytical Methods

The aggregate size distribution of bulk samples was assessed using the standard dry-sieving procedure [14]; 100 g of bulk soil was shaken automatically for 10 min and divided into 6 particle size levels using 53, 250, 1000, 2000, and 4000 μm sieves. The distribution of each particle size fraction was calculated by determining the weight of each size fraction.

The pH was measured using the ion selective electrode method [14]. TOC was measured using the volumetric method [14]. In brief, 10 mL of 1.0 mol L^{-1} of $K_2Cr_2O_7$ and 20 mL of concentrated H_2SO_4 were added into a volumetric flask containing 0.5 g of sample. After 30 min, the mixtures were titrated with 0.5 mol L^{-1} of Fe_2SO_4 with a few drops of phenanthroline as the indicator.

For each particle size fraction and bulk sample, 0.1 g fine soil was mixed with 5 mL HNO_3, 2 mL $HClO_4$, and 1.5 mL HF in PTFE reactor. Closed digestion was maintained for 6 h at 160 °C using a drying oven (DGB/20-002, Chongqing experimental equipment factory, China). The total amounts of the HMs As, Cr, Pb, and Cd were determined by ICP-MS, (Thermo-X7, Thermo Scientific, Waltham, MA, USA). In the process of analysis, Tibetan standard soil (GBW08302, National Institute of Metrology, Beijing, China) was used as the quality control sample. The recoveries of Cr, As, Cd, and Pb were 86–96%, 102–110%, 90–123%, and 99–103%, respectively. The relative standard deviations (RSDs) of all HMs for blank samples and duplicate samples were less than 5% and 10%, respectively. The analytical batch with ten samples included one standard to control the quality and the blank value was subtracted during data processing.

The leaching test was carried out with 10 g of sample in a conical bottle and 100 mL of high purity water with a pH of 5.0 ± 0.2 was added. The mixed liquid was stirred using a magnetic agitator (HJ-6A, Changzhou Guohua Electric Appliance Co., Ltd., Changzhou, China) for 8 h, rested for 16 h, then centrifuged and filtered with a 0.45 μm filter. The supernatant was separated from the solid phase. ICP-MS was used to determine the HMs in the filtrate. A batch consisted of six samples which included one duplicate sample and one blank sample. The RSDs of duplicate samples were generally less than 10%.

2.3. Assessment Factors

2.3.1. Distribution Factor (DFx)

In order to estimate which particle size fractions of HMs were preferentially enriched, the distribution factor (DF$_x$) was calculated using the following Equation [14]:

$$\mathrm{DF}_x = X_{fraction}/X_{bulk} \tag{1}$$

where X_{bulk} and $X_{fraction}$ are concentrations (mg kg^{-1}) of HMs in bulk and a given fraction sample, respectively. If DF$_x$ > 1, the HMs are assumed to be accumulated in this fraction.

2.3.2. Mass Loading of HMs

Metal mass loading is one of the most important pollution indexes, which was used to evaluate the mass loadings of HMs in each particle size fraction [14]. In order to compute HM mass loading (%) on a grain size fraction basis (GSF$_{loading}$), the following equation was used [36]:

$$\mathrm{GSF}_{loading} = (\mathrm{HM}_i \times \mathrm{GS}_i) / (\sum_{i=1}^{6} (\mathrm{HM}_i \times \mathrm{GS}_i) \times 100 \tag{2}$$

where HM$_i$ is the HM (i) content (mg kg^{-1}) in an individual grain size fraction and GS$_i$ is the mass percentage of an individual fraction, which has limits of 0–100%. The summation of GSF$_{loading}$ for the individual samples will always be 100%.

2.3.3. Pollution Index

The single pollution factor index (Pi) and integrated pollution index (IPi) were used to evaluate the environmental quality of the different particle size fractions of paddy soil. The calculation of Pi can be expressed as [37]:

$$\mathrm{P}i = \mathrm{C}i/\mathrm{S}i \tag{3}$$

and the mathematical formula of the IPi is [37]:

$$\mathrm{IP}i = \sqrt{\frac{\left(\frac{1}{n}\sum_{i=1}^{n}\mathrm{P}i\right)^2 + [\max(\mathrm{P}i)]^2}{2}} \tag{4}$$

where Pi is the pollution index of HM$_i$, Ci (mg kg^{-1}) is the actual concentration of HM$_i$, and Si is the reference content (mg kg^{-1}) of HM. In this study, the mean contents of HMs in all uncultivated soils were employed as reference concentrations. The Pi and IPi values were classified as follows: clean ($\leq$1), slightly polluted (1–2), moderately polluted (2–3), and heavily polluted ($\geq$3) [38].

2.3.4. Leachability

To evaluate the potential migration of HMs absorbed in wet soil, a leachability experiment was conducted. The leachable proportion of water (WLP, %) for HMs in different particle size fractions was calculated using following equation:

$$\mathrm{WLP} = \mathrm{L}_i/\mathrm{X}_i \times 100 \tag{5}$$

where L$_i$ and X$_i$ are leaching contents and total concentrations of HMs in a given particle size fraction, respectively.

2.4. Statistical Analysis

The analytical data were expressed using arithmetic means. All analyzed data were standardized using *t*-test transformation. The data analysis was performed using Excel 2019 software (version number:2305 Build 16.0.16501.20074) and Origin 2021 was used to draw the maps.

3. Results and Discussion

3.1. Soil Properties and Its Particle Size Fractions

The physical and chemical properties of paddy soil samples from three sites are shown in Table 1. The soils were weakly acidic with a mean pH of 6.14 ± 0.37. The surface soils were characterized by the largest proportion of the coarsest aggregates, i.e., >4000 μm (20.21–30.59%). The finest aggregates of <53 μm particles were 5.65%, 17.28%, and 11.95% in Feidong soil, Shouxian soil, and Shucheng soil, respectively. Soil microaggregate particles can easily carry HMs to the atmosphere or migrate to deep soil and groundwater under the influence of rainwater and cultivation; therefore, they have stronger environmental hazards and should be given sufficient attention [31,39].

The contents of TOC were 12.59 g kg^{-1}, 13.26 g kg^{-1}, and 13.22 g kg^{-1} in Shouxian soil, Shucheng soil, and Feidong soil, respectively. A similar finding was reported in Anhui Province with a 12.6 g kg^{-1} mean concentration [40]. In our study, the TOC concentrations (Table 1) increased from coarse to fine particle size fractions, and the smallest particle size aggregates (<53 μm) had the largest concentrations with 19.75, 16.96, and 16.18 g kg^{-1} in Shouxian soil, Shucheng soil, and Feidong soil, respectively.

Sorption by iron oxides and manganese oxides has been identified as one of the major mechanisms by which HMs are retained in soil [41,42]. The concentrations of Fe and Mn were 17.64 and 0.26 g kg^{-1} in Shouxian soil, 9.31 and 0.10 g kg^{-1} in Shucheng soil, and 11.33 and 0.09 g kg^{-1} in Feidong soil, respectively. For both Fe and Mn, the highest contents were found in the finest particle size fractions with an increasing trend from coarse to fine particle size.

Table 1. Properties of paddy soil samples in Anhui Province.

	pH	Particle Size Fractions, %	TOC	Fe	Mn	Cr	As	Cd	Pb
			g kg^{-1}			mg kg^{-1}			
Shouxian									
bulk			12.59	17.64	0.26	34.05	7.34	0.15	13.80
<50 μm		5.65	19.75	22.71	0.33	42.95	9.35	0.19	20.28
50–250 μm		3.50	11.57	18.76	0.31	35.40	8.20	0.15	16.32
250–1000 μm	6.54	23.20	13.26	17.48	0.29	34.68	7.49	0.17	15.57
1000–2000 μm		19.99	11.78	15.90	0.23	31.29	6.94	0.13	11.91
2000–4000 μm		17.07	11.33	17.40	0.25	33.48	7.13	0.15	13.42
>4000 μm		30.59	12.46	17.97	0.25	33.91	7.09	0.15	12.40
Feidong									
bulk			13.26	9.31	0.10	21.52	10.01	0.15	14.98
<50 μm		17.28	16.96	13.16	0.14	29.39	11.97	0.20	19.15
50–250 μm		14.59	16.44	10.32	0.11	21.95	10.50	0.15	14.86
250–1000 μm	5.82	16.72	13.07	8.64	0.09	19.39	10.34	0.13	15.09
1000–2000 μm		15.58	12.48	8.12	0.09	20.90	10.16	0.16	15.12
2000–4000 μm		15.62	12.10	8.59	0.09	19.52	9.46	0.13	13.36
>4000 μm		20.21	10.75	7.33	0.07	18.26	8.60	0.12	12.56

Table 1. *Cont.*

	pH	Particle Size Fractions, %	TOC	Fe	Mn	Cr	As	Cd	Pb
			g kg⁻¹			mg kg⁻¹			
Shucheng									
bulk			13.22	11.33	0.09	19.58	11.11	0.17	15.93
<50 µm		11.95	16.18	15.19	0.12	27.81	13.89	0.21	22.27
50–250 µm		18.76	12.29	10.54	0.08	18.22	10.11	0.15	17.06
250–1000 µm	6.07	16.83	12.77	11.73	0.10	20.52	11.79	0.17	17.57
1000–2000 µm		14.95	13.45	11.88	0.11	22.05	10.32	0.18	18.31
2000–4000 µm		14.84	13.14	7.69	0.07	16.11	9.98	0.12	12.67
>4000 µm		22.69	13.82	11.67	0.08	16.31	10.37	0.17	11.00
reference value [a]						21.43	7.93	0.06	12.75
	pH ≤ 5.5					150	40	0.3	70
Threshold values [b]	5.5 < pH ≤ 6.5					150	40	0.3	90
	6.5 < pH ≤ 7.5					200	30	0.3	120
	pH >7.5					250	25	0.6	170

[a], Mean value of HMs in all bulk uncultivated soils; [b], the risk screening values for soil pollution (soil environmental quality risk control standard for soil contamination of agricultural land in China (GB 15618-2018) [43]).

3.2. HM Enrichment and Particle Size Distribution

The mean contents of Cr, As, Cd, and Pb in the three paddy soils were 25.05, 9.49, 0.16, and 14.90 g kg⁻¹, respectively, which lower than the risk screening values for soil pollution (soil environmental quality risk control standard for soil contamination of agricultural land (GB 15618-2018)). The mean values of Cr, As, Cd, and Pb in the three uncultivated soils were 21.43, 7.93, 0.06, and 12.75 g kg⁻¹, respectively (Table 1).

Different particle size fractions have various compositions and properties, which constrain the behavior of pollutants in the soil microenvironment. It is very important to explore the partitioning of soil particle size to evaluate the mobility and bioavailability of HMs [44]. The distribution factors (DF_x) of Cr, As, Cd, and Pb are shown in Figure 2. Generally, the HMs in the three paddy soils display similar patterns, i.e., the finest fractions (<53 µm) had the highest mean DF_x values for Cr (1.35), As (1.25), Cd (1.28), and Pb (1.38) in all the paddy soil samples, and the coarse particle size fractions (>2000 µm) had the lowest DF_x values of Cr (0.89), As (0.92), Cd (0.86), and Pb (0.81). A few other studies have also found the preferential enrichment of Cd, Pb, and Cr in the finer aggregate [13,31,44].

Generally, the accumulation of HMs in all paddy soils increased with decreasing particle size. Such a pattern is usually attributed to an increase in the specific surface area and the presence of clay minerals, organic matter, and Fe/Mn/Al oxides in the microaggregate [24,30,36,44]. Meanwhile, HMs were also enriched in (DF > 1) the larger particle size fractions such as Cd and Pb in 250–1000 µm fractions in Shouxian soil, Cr and Cd in 1000–2000 µm fractions and Pb in 250–2000 µm fractions in Shucheng soil, and As in 250–1000 µm fractions and Cd in 1000–2000 µm fractions in Feidong soil. HMs accumulated in the fine as well as the coarse aggregates, which may be due to different complex factors, including new HM input firstly adsorbed by the coarse aggregates, and microaggregates embodied in the coarse particles [45].
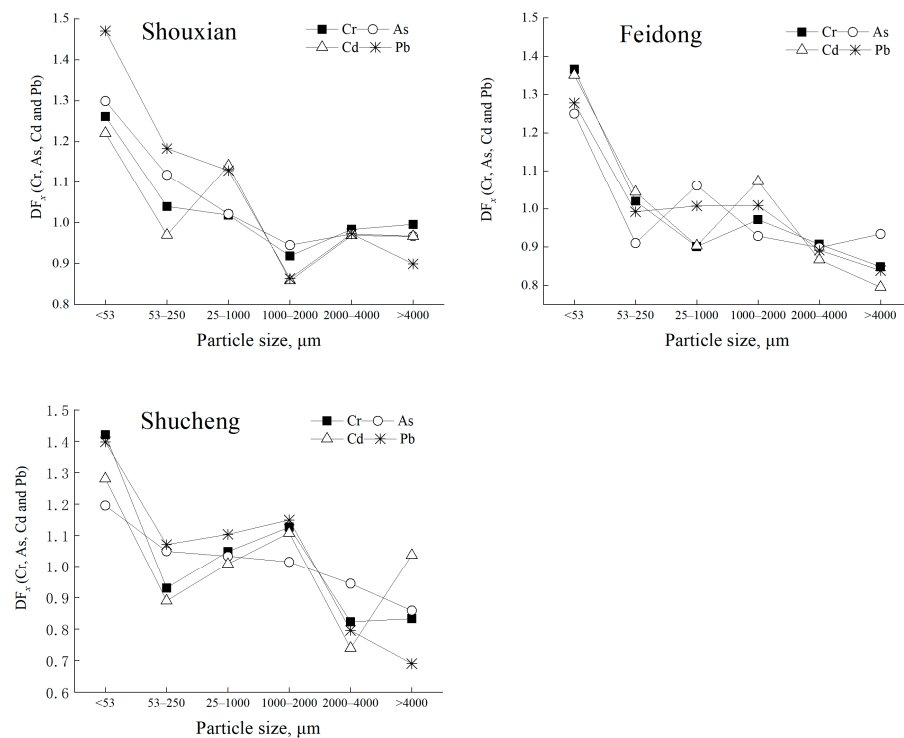
Figure 2. Distribution factors of HMs in the different soil particle size fractions of Shouxian, Shucheng, and Feidong.

3.3. Mass Loading and Pollution Assessment

In order to estimate the contribution of each particle size fraction to the concentrations of HMs in bulk soil, HM mass loading was calculated, and the results are shown in Figure 3. Generally, the highest contributions of Cr (22.17%), As (22.64%), and Cd (23.26%) came from the coarsest fractions (>4000 μm) and the highest contributions of Pb (20.53%) came from the 250–1000 μm fractions. In the coarsest fraction, organic carbon (OC) was easily decomposed, and the adsorbed pollutants were released or redistributed as the OC decomposed [46]. However, microaggregate (<250 μm) contributions of Pb, As, Cd, and Cr were 28.60%, 26.70%, 26.97%, and 27.89%, respectively, and, for Cr, As, Cd, and Pb, the mass loadings of <53 μm particles were up to 15.90%, 14.41%, 15.21%, and 15.70%. It should be noted that microaggregates, especially <53 μm, were easily transferred into deep soil, groundwater, or resuspended to the atmosphere as well, which could result in potential contamination in the future [30].
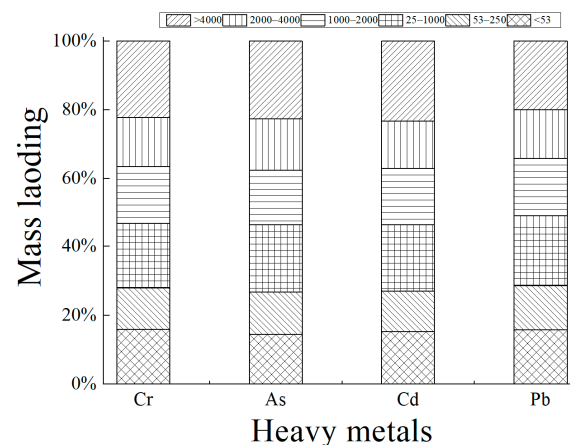

Figure 3. HM mass loadings of the six soil particle size fractions.

In order to evaluate the contamination levels of HMs in the different soil particle size fractions, the P*i* and IP*i* were computed and are depicted in Figure 4. The pollution levels of Cr, As, and Pb in the six aggregates with different particle sizes were clean (P*i* ≤ 1) or slightly polluted (1 < P*i* < 2). However, the pollution levels of Cd in aggregates with particle sizes <53 μm, 53~4000 μm, and >4000 μm were heavily polluted (P*i* = 3.17), moderately polluted (P*i* = 2.11–2.51), and slightly polluted (P*i* = 1.75), respectively. The highest IP*i* was 2.61 (moderately polluted) which came from the <53 μm particle size aggregate, the second IP*i* (2.07) was in the 250–1000 μm fraction, and the lowest IP*i* (1.49) was in the >4000 μm fraction. Generally, the finest (<53 μm) had the highest pollution levels (P*i* and IP*i*) for all HMs, and the pollution levels decreased with increasing particle size. This means that HMs artificially introduced from agricultural activities tend to be adsorbed in small particle size soil aggregates. Related studies have pointed out that due to the large specific surface area of small particle size soil aggregates and the existence of a negative charge, some HMs were finally protected and difficult to be consumed [14].
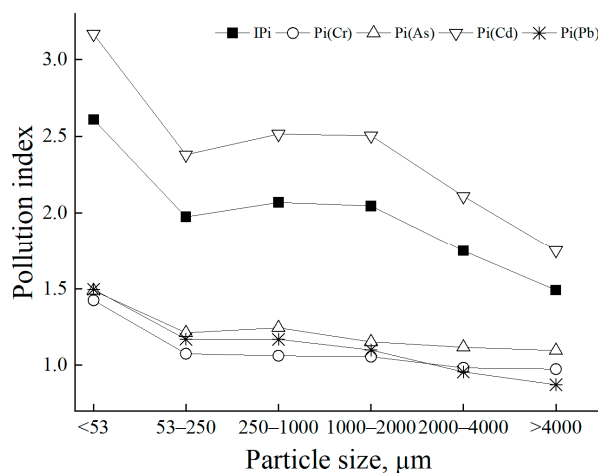

Figure 4. Single-factor index of HMs and integrated pollution index values of the six soil particle size fractions.

3.4. Leaching Test

To check the mobility of the HMs in different aggregates, a leaching test was performed [47] and the results are shown in Figure 5. In order to eliminate the effects of the various contents in different aggregates, the data were normalized to the total concentrations of each aggregate.
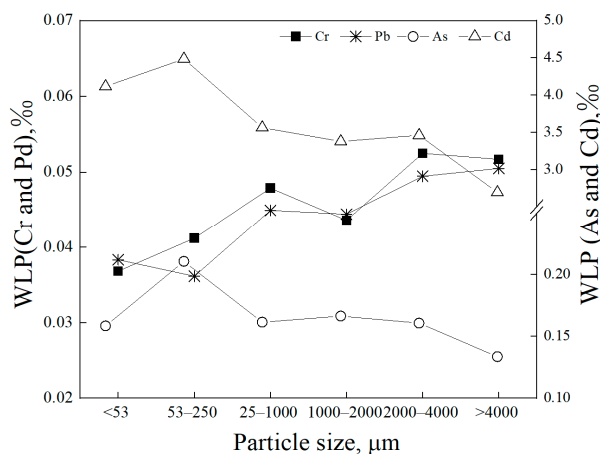

Figure 5. Leachability of HMs in the different aggregates.

The leachable proportions of HMs in various particle size fractions were below 5‰, which were less than the leachability of contaminated soils and urban soils studied by Abollino [47] and Wang [48], respectively. Abollino's study showed that the leachability behaviors of Cr, Cd, and Pb were 0.108%, 5.665%, and 0.390%, respectively, while Wang's study showed that the leachability behaviors of Cr, Cd, Pb, and As were <0.015%, 4%, <4.5%, and 2.17%, respectively. The leachability of As and Cd was higher than Cr and Pb by from one to two orders of magnitude. For Cr and Pb, the lowest leachability was found in the finer microaggregates (<250 μm), and leachability increased with decreasing aggregate size; the coarser (>2000 μm) particle size fractions had the highest values of Cr (0.053‰) and Pb (0.051‰). The trend of HMs adsorbed in the finer fractions may be due to the fact that the HMs in fine fractions were stronger bound with the OC [49] and mineral surface [50]. It is well known that iron oxide particles are effective sorbents for HMs [51]. However, the opposite distribution mode for As and Cd, with the highest leaching of 0.21‰ and 4.48‰, were found in the <250 μm particle size fractions, the coarsest fractions (>4000 μm) had the lowest values of 0.13‰ for As and 2.69‰ for Cd, respectively. This is means that HMs of As and Cd in the <250 μm particle size fraction had the larger risk for groundwater and surface runoff.

Table 2 shows the correlation analysis results among HMs with TOC, Fe, and Mn contents. There were positive correlations (R^2 = 0.60–0.84) between Pb, Cd, As, Cr and TOC in Shouxian soil, and, between Cr, Cd, Pb and TOC in Feidong soil; however, between Cr, Pb, As, Cd and TOC in Shucheng soil and between As and TOC in Feidong soil there were weak positive correlations (R^2 = 0.24–0.49), which suggest that the particle distribution of TOC has a strong influence on the distribution of these metals. Similar conclusions have been made by other studies [45,49]. For example, Li's study found that the correlation coefficients R^2 for Cr and Pb with TOC in soil aggregate particles were 0.5846 and 0.5959, respectively [45]. As shown in Table 2, there were positive correlations between Fe (R^2 = 0.51–0.97) and Mn (R^2 = 0.49–0.96) contents in all paddy soils and As, Pb, Cd, and Cr.

Table 2. The correlation coefficient (R^2) values between As, Pb, Cd, and Cr concentrations and TOC, Fe, and Mn contents of the soil particle size fractions.

		TOC	Fe	Mn
Shouxian	Cr	0.84	0.97	0.69
	As	0.68	0.89	0.81
	Cd	0.60	0.52	0.55
	Pb	0.62	0.79	0.91
Feidong	Cr	0.60	0.90	0.96
	As	0.32	0.51	0.49
	Cd	0.60	0.75	0.89
	Pb	0.55	0.77	0.86
Shucheng	Cr	0.46	0.67	0.87
	As	0.28	0.27	0.53
	Cd	0.49	0.94	0.63
	Pb	0.24	0.39	0.77

4. Conclusions

This study researched the distribution characteristics of HMs in aggregates of different soil particle size fractions of a paddy field located in Hunan Province, China. The mean concentrations of Cr, As, Cd, and Pb in all the paddy soil aggregates were lower than the risk screening values for soil contamination but higher than the mean values of Cr, As, Cd, and Pb in uncultivated soil, meaning that the paddy soils were polluted by Cr, As, Cd, and Pb. The distribution of HMs in different particle size fractions increased with decreasing particle size. Although the leachability of HMs in microaggregates was very low, the high mass loading and mobility of microaggregates still require attention during the

environmental risk assessment. Soil is a very complex heterogeneous medium on the Earth, and the formation of soil aggregates is the comprehensive result of physical, chemical, and biological processes in soil and human activities. HMs can be strongly fixed in soil aggregates and then can migrate into the environment in various ways, including being absorbed by plants, washed by water, washed by surface runoff, or transported with lost particles in various erosion processes. Aggregate size plays an important role in controlling the distribution, availability, and migration of heavy metals in soil.

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