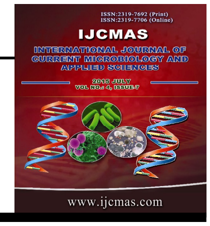

International Journal of Current Microbiology and Applied Sciences
ISSN: 2319-7706 **Volume 4 Number 7 (2015) pp. 65-75**
http://www.ijcmas.com

Original Research Article

Characterization of the Particle Size Fraction Associated Heavy Metals in Arablesoils from Ahwaz size, Iran

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Keywords

Particle size fraction;
Heavy metal
Arable soil;
Leachability

A B S T R A C T

Arable soils were sampled from Ahwaz city. The particle size fractions associated Cr, As, Cd and Pb were characterized and their leachability was analyzed as well. The pollution of heavy metals in the arable soils was not severe except Cr (204 mg kg^{-1}) in Haikou. The distribution of heavy metals increased with decrease of particle size. Although the smallest fractions (b53μm) occupied only 5.08–9.57%, they had the highest distribution factor (DF) of 3.50 for Cd, 2.11 for Pb, 1.73 for Cr and 1.09 for As, respectively. The contributions of micro-aggregates (<250μm) to the total amount of heavy metals was 41.9% for Cr, 44.6% for As,61.6% for Cd and 48.6% for Pb, respectively, and the second mass loading came from the particles of 250–1000μm. The residues of Cr, Cd and Pb were correlated positively with the contents of organic carbon as well as Fe in fractions, while a large variation distribution of As was found in particles, indicating its high ac-tivity in soil microenvironment. The lowest leachability was found in the easily migratory micro-aggregates, which should be taken into account in the future environmental risk assessment and soil remediation.

Introduction

Heavy metals in soil have attracted extensive attention worldwide since most of them are harmful to the crop quality and pose a potential threat to human health through the food chain (Huang and Jin, 2008; Quinton and Catt, 2007). In China, the harsh fact is that soil pollution by heavy metal is getting worse with the rapid urbanization for decades (Wei and Yang, 2010). Understanding the distribution characters of heavy metals in soil is the premise of risk assessment and soil remediation. Numerous studies have been conducted to determine heavy metals in both bulk soils and different particle size fractions. However, most previous studies are focused on urban soil, gar-den soil, dust and sediment (Acosta *et al.*, 2009, 2011; Ajmone-Marsan *et al.*, 2008; Beamer *et al.*, 2012; Sutherland, 2003) and only few data are available about the distribution of heavy metals in various particle size fractions from arable soil (Qian *et al.*, 1996; Quenea *et al.*, 2009; Zhang *et al.*, 2003). The mobility and biological effectiveness of soil heavy metals have a strong correlation with the size and composition of particle fractions (Zhang *et al.,* 2003). Various size fractions have different composition and properties, which affected the behavior of pollutants in the soil microenvironment (Acosta *et al.,* 2011). Generally, fine particles have a higher ability to carry the heavy metals due to the increase of specific surface area, the presence of clay minerals, organic matter, and Fe/Mn/Al oxides in the micro-aggregate (Cai *et al.,* 2002; Ljung *et al.,* 2006; Semlali *et al.,* 2001; Sutherland, 2003; Wang *et al.,* 2006), which would protect the heavy metals from remedial attempts. Moreover, since the fine soil fractions are often preferentially transported to deep soil, surface/ground water and air, they are more harmful to the environments (Farenhorst and Bryan, 1995; Uusitalo *et al.,* 2001). Therefore, exploring the partition in soil particle sizes was very important for the assessment of mobility and bioavailability of heavy metals in soil (Wang *et al.,* 2006).

Ahwaz city is located in the South west Iran and it is becoming an international travel insight according to the national developing plan. The industrialization caused heavy metal accumulation in arable soil. The concentration of Cr in the suburban soil in Ahwaz was up to 586.70 mg kg^{-1}. The rich precipitation and strong biological action in the study area could result in a rapid cycle of organic carbons in soil. The distribution character of heavy metals in particle size fractions is still unknown in arable soils in Ahwaz. Moreover, in order to predict both potential mobility and their bioavailability to plants, it is necessary to assess the leachability of heavy metals in different particles. Up to now, only few data about water-leaching risk of heavy metals in different particle size fractions have been reported (Magnuson *et al.,* 2002; Zhang *et al.,* 2003). In order to assess the environmental risk and take applicable measures in the future, it is important to know in which scale particles the heavy metals are preferred to distribute. The objectives of this study are to: (1) assess the contamination levels of toxic Cr, Pb, As and Cd in arable soils in Ahvaz city; (2) investigate the distribution patterns and leachability of heavy metals in different particle size fractions; and (3) discuss the factors effecting the residues of heavy metals indifferent particle size fractions.

Study area

The industrial town of Ahwaz is situated in SW of Iran Capital of Khuzestan province, Iran, occupying an area of 815.85 km2 (0.315 mile2) (Fig. 1). The climate of the study area which is located in Khuzestan Province, almost near Persian Gulf is arid and sultry and almost cold and rainy in winter. Average temperatures in the study area are 23 °C (73 °F) in January, 38 °C (100 °F) in April and 49 °C (120 °F) in July. The rainy season normally extends from late December to almost end of March with an average annual rainfall of 213 mm year-1.

The Altitude datum in industrial town is about 25 m above the sea level. It is surrounded by open arid areas and a semi forest palm plantation in the south. The prevailing wind direction is from northwest to southeast.

Based on Standard US Department of Agriculture Soil Textural Classification Triangle, the soil texture in the study area was classified as clay loam to silt loam, with a lesser extent of sandy loam and loamy sand. The pH values ranging in narrow interval from 8.1 to 8.2, which suggests sub-alkaline condition for all the soil samples.

Sampling and analysis

39Surface sediment samples were collected at between February and January 2012. The samples collected from each site consisted of 4–5 composite samples. Composite sediments (top 5 cm of surface) were taken by using a self-made sediment sampler . After sampling, the sediment samples were sealed in clean polyethylene bags, placed in a cooler at 4°C, and transported to the laboratory immediately for further analysis. Samples were digested in aqua regain; 1 g sample in 3 ml 3:1:2 HCl:HNO3:H2O, analyzed using inductively coupled plasma optical spectrometry emission technique (ICP-OES) at the Geological Survey of Finland. As, Cd and Hg were determined by GFAAS. The detection limits for the elements (in mg/kg) were: As (0.1), Cd (0.01), Cr (1), Cu (1), Hg (0.01), Ni (2), Pb (5) and Zn (1). The two digestion methods that were used in 1994 and 2004 are reasonably similar in strength and comparable results should generally be expected, when samples are analyzed in one batch.

Assessment methods

Enrichment factor (EFx): The enrichment factor was used to assess the anthropogenic ally introduced heavy metal, and it was calculated by Eq. (1) (Chan *et al.*, 2001):

$$EF_x = X/X_{ref}. \qquad (1)$$

Where Xis the concentration in soil (mg kg^{-1}) and X_{ref} is the reference concentration (mg kg^{-1}). In this study, the contents of heavy metals in the uncultivated soils were used as background reference. The EF was classified as follows: >16.0 (excessive), 16– (moderate) and 2.0–1.1 (slight) 8.1 (very severe), 8.0–4.1 (severe), 4.0–2.1) (Acosta *et al.*, 2011).

Distribution factor (DFx)

In order to estimate in which size fraction the heavy metals are preferentially enriched, distribution factor (DFx) was calculated by Eq.(2)(Acosta *et al.*, 2009):

$$DF_x = X_{fraction}/X_{bulk}. \qquad (2)$$

Where, X fraction and X bulk are contents (mg kg^{-1}) of heavy metal in a given fraction and bulk sample, respectively. The heavy metal is assumed to be accumulated in this fraction, if DFx>1.

Mass loading

Another important index to assess the contamination of heavy metals is the mass loading. Loading combines heavy metal concentrations, on a grain size basis, with data on the mass percentage. The index was calculated by Eq. (3) (Sutherland, 2003):

$$GSF_{loading} = (HM_i \times GS_i)/\left(\sum_{i=1}^{6}(HM_i \times GS_i) \times 100\right). \qquad (3)$$

Where, HMi is the heavy metal content (mg kg^{-1}) in the individual grain size fraction (i) and GSiis the mass percentage of the individual fraction.

Leachability

The water-leachable proportion (WLP, %) of heavy metal in each size fraction was calculated as described in Eq. (4):

$$WLP = L_i/X_i \times 100. \qquad (4)$$

Where L_i and X_i are leaching concentrations and the total contents of heavy metals in a given particle size fraction, respectively.

Statistical analysis

Statistical analysis was performed using Excel 2007 (Microsoft Co., USA) and the figures were drawn by Origin 7.0 (Microcal Co., USA).

Results and discussion

Soil properties

The soil properties and content of heavy metals in arable soils are shown in table 1. The surface soils were characterized with a dominant fraction of fine aggregates of 53–250μm (32.4–35.4%) and coarse aggregates of 250–1000μm (31.4–38.0%). The contents of TOC in bulk soils were 24.0 g kg^{-1} in Haikou, which was similar with the mean level in northeastern China (22.90 g kg^{-1}) (Liang *et al.*, 2009). While the content of TOC in arable soil from Ahwaz and Abadan was pretty low with only 6.25 g kg^{-1} and 3.62 g kg^{-1}, respectively. In terms of the distribution of particles, the contents of TOC increased with the decrease of particle sizes, and the smallest fraction (<53μm) had the highest organic carbon contents of 25.4, 17.5 and 11.1 g kg^{-1} in Haikou, Qionghai and Tunchang soils, respectively. Organic carbons in finer fractions were biologically stable, and heavy metals in this fraction were considered more persistent and harmful to the environment (Abdulfatah *et al.*, 2009; Acosta *et al.*, 2011; Semlali *et al.*, 2001).

In addition, another major mechanism of heavy metals retained in soil was related to iron oxides (Kahapanagiotis *et al.*, 1991). The concentrations of Fe were 50.2 g kg^{-1} in Haikou and only 9.59 and 12.2 g/kg^{-1}in Ahwaz and Abadan soils, respectively. The correlations between heavy metals and TOC as well as Fe content were discussed later.

Heavy metal enrichment and particle size distribution

As presented in table 1, the concentrations of Cr (204 mg kg^{-1}) in arable bulk soil from Ahwaz exceeded the limit (150 mg kg^{-1}) for the protection of agriculture according to the Environmental Quality Standard for Soils in Iran. The contents of Cr, As, Cd and Pb in other samples were lower than the allowable concentrations, which indicate no harmful effect on the normal agriculture at the present level. The arable soils studied were slightly or moderately polluted by the heavy metals with EF of 1.23–1.95 for Cr, 1.18–2.67 for As, 1.21–1.55 for Cd and 1.01–2.08 for Pb, respectively. The distribution factors (DFx) of Cr, As, Cd and Pb are shown in figure 2. Generally, the distribution of heavy metals in soils increased with decrease of particle size, e.g. the smallest fractions (<53μm) had the highest mean DF of 3.50 for Cd, 2.11 for Pb, 1.73 for Cr and1.09 for As, respectively. A few other studies also reported similar pat-terns (Acosta *et al.*, 2011; Beamer *et al.*, 2012; Sutherland, 2003; Zhang *et al.*, 2003). The lowest DF was found in the coarse particles (>1000μm) with DF of 0.79 for As, 0.80 for Cd, 0.92 for Pb and 0.96for Cr. Quenea *et al.* (2009) reported that contents of Cd in fine fractions (20–50μm) was about 67 times those in >200μm size fractions in agriculture soils, and about 375 times in fine particles as those in coarse one in another study (Harter and Naidu, 1995). In this study, the concentrations of Cd (0.64 to 0.96 mg kg^{-1}) in <53μm fractions were 4.5–11.0 times higher than those in coarse particles. A larger variation distribution of As in particles was found in this study, which indicates that As is active in the soil microenvironment and it is easy to migrate between particles.

Meanwhile, we noticed that larger size fractions could enrich the heavy metals as well, e.g. Cr in 250–1000μm fraction, Pb in >4000μmfraction in Ahwaz and Abadan, and Cd in 2000–4000μm fraction in Haikou, respectively. The fact that heavy metals accumulated in not only fine but also coarse particles was probably due to different complex factors, including new input of heavy metals firstly adsorbed by the coarse particles, and micro-aggregates embodied in the coarse aggregates and so on (Li *et al.,* 2001, 2008a; Qian *et al.,* 1996).

Mass loading and leachability

In order to estimate the contribution of fractions to the total contents of heavy metals in bulk soil, mass loading was calculated as shown in figure 3. The micro-aggregates (<250μm) contributed to the total contents of 41.9% for Cr, 44.6% for As, 61.6% for Cd and 48.6% for Pb, respectively. The mass loading of <53μm particles was up to 22.7% for Cd. It should be noted that the micro-aggregates especially53μm were easily transferred and could induce further contamination (Ljung *et al.,* 2006; Semlali *et al.,* 2001). The second mass loading came from the particles of 250–1000μm (34.6% for Cr, 38.6% for As,19.3% for Cd and 31.4% for Pb, respectively), in which the organic car-bon was easily decomposed and the adsorbed pollutants would be re-leased or reassigned in the bulk soil (Pichler *et al.,* 1996). The lowest loading (8.52% for Cr, 6.68% for As, 10.6% for Cd and 8.17% for Pb respectively) came from the coarse aggregates (>2000μm). The tendency of heavy metals being adsorbed in the finer fractions should be taken into account during the environment risk assessment and soil remediation. In order to check the leachability of heavy metals in different particles, the leaching test was necessary and the results were plotted in figure 4.

The data were normalized to the total contents of each particle to eliminate the effects of the various concentrations in different particles. The leachable proportions of heavy metals in various size fractions were pretty low, which were 0.07‰ for Cr, 0.37‰ for As, 1.45‰ for Cd and 0.05‰ for Pb. The results in this study were much less than those in contaminated soils studied by others, which was 0.11% for Cr,5.67% for Cd and 0.39% for Pb (Abollino *et al.,* 2002), and <0.02% for Cr, 4.00% for Cd,<4.50% for Pb and 2.20% for As, respectively (Wang,2009). The lowest leachability of Cd, Cr and Pb was found in the micro-aggregates (<250μm) while higher leachability was found almost in coarser fractions. For As, the highest leaching was found in the b53μm particles. The low leachability of heavy metals in finer fractions may be due to the fact that the heavy metals in these fractions were strong bounded with the organic carbon (Quenea *et al.,* 2009) and the mineral surface (Trivedi and Axe, 2001). Moreover, it was well known that iron oxide particles were effective sorbents for heavy metals (Nachtegaal and Sparks, 2004).

In order to explore the effects of the organic carbon and iron content on the adsorption of heavy metals, the relationships are shown in figure 5. There were positive correlations with TOC as well as with Fe in all soils for Cr, Pb and Cd in different size fractions, which suggested that the particle distribution of TOC and iron oxide strongly influenced the distribution of these heavy metals. A few studies drew the similar conclusion (Li *et al.,* 2001; Quenea *et al.,* 2009). However, the correlations for As were obviously different with a correlation (R^2=0.39) in Ahwazand a weaker correlation (R^2=0.10) in Abadan, while a negative correlation was found in Qionghai. Ljung (Ljung *et al.,* 2006) also found that As content had no correlations with OC content. The correlation value between As and iron was negative in Ahwaz soil. The different patterns with the TOC and iron may result in the transferring for As in the microenvironment.

The arable soils in Hainan Island have been polluted by Cr, As, Cd and Pb. The distribution of most heavy metals in different size particles was increased with decreasing particle size. The content of organic carbons as well as Fe played an important role in the distribution of Cr, Cd and Pb, while As had a different pattern. The mass loading of heavy metals in micro-aggregates was high while their leachability was very low, which should be paid attention during the environment risk assessment and soil remediation.

Table.1 Soil properties and contents of heavy metals in both bulk and their particle size fractions of the studied soils

	Particle size fractions, %	TOC g kg^{-1}	Fe g kg^{-1}	Cr	As	Cd	Pb
				mg kg^{-1}			
Haikou							
Bulk		24.0	50.2	204	3.77	0.25	19.3
<53 μm	5.08	25.4	69.2	228	4.38	0.64	23.6
53–250 μm	32.4	23.5	51.1	198	4.20	0.24	22.1
250–1000 μm	31.4	23.9	44.7	214	4.06	0.16	21.9
1000–2000 μm	16.6	23.0	47.3	210	3.37	0.14	14.2
2000–4000 μm	8.57	23.0	44.1	182	1.82	0.39	15.0
>4000 μm	5.91	22.5	45.4	182	3.30	0.20	14.7
Qionghai							
Bulk		6.25	9.59	21.2	2.49	0.20	17.2
<53 μm	5.12	17.4	19.1	46.2	1.20	0.96	59.0
53–250 μm	35.4	7.96	12.4	23.1	3.89	0.39	26.8
250–1000 μm	33.5	3.67	10.1	22.1	3.14	0.12	16.6
1000–2000 μm	17.9	3.29	7.73	25.3	1.60	0.11	14.7
2000–4000 μm	5.45	11.4	9.63	14.9	2.89	0.24	14.6
>4000 μm	2.61	5.20	11.8	20.3	2.45	0.29	21.4
Tunchang							
Bulk		3.62	12.2	23.3	4.49	0.21	19.4
<53 μm	9.57	11.1	16.4	43.9	7.29	0.66	32.6
53–250 μm	34.3	4.21	12.4	20.5	6.65	0.16	16.2
250–1000 μm	38.0	1.69	13.8	26.5	8.40	0.10	16.7
1000–2000 μm	11.0	3.29	13.4	20.4	3.61	0.13	19.6
2000–4000 μm	4.19	1.84	12.8	23.3	4.67	0.14	19.1
>4000 μm	2.93	2.68	11.0	26.9	1.05	0.06	20.0

Fig.1 Location map of study area

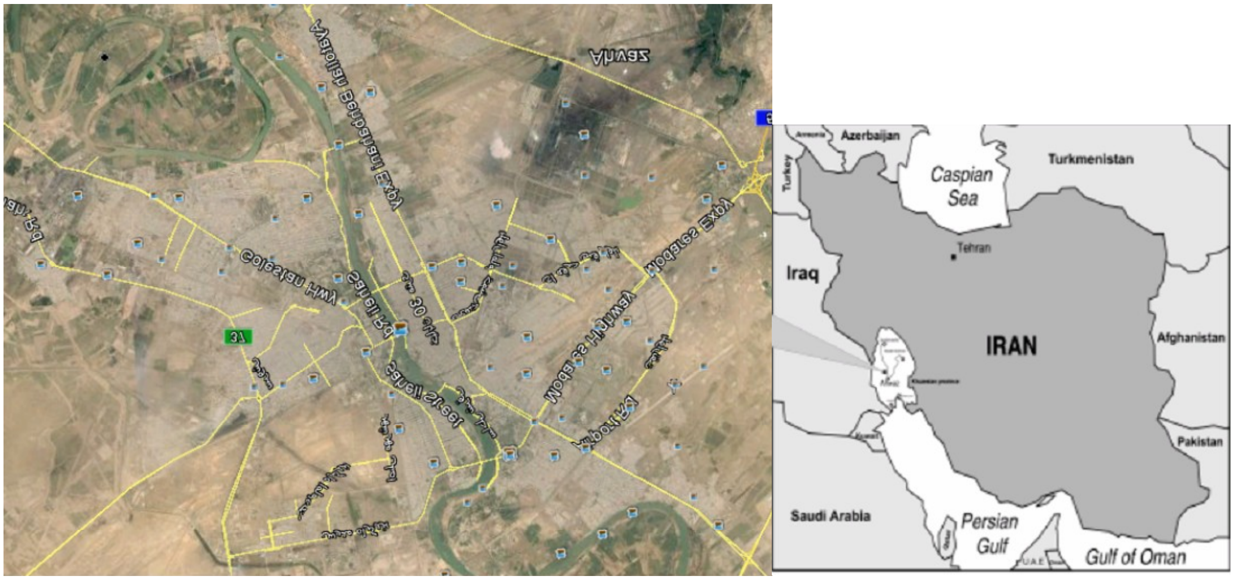


Fig.2 Distribution factors of heavy metals in different particle size fractions

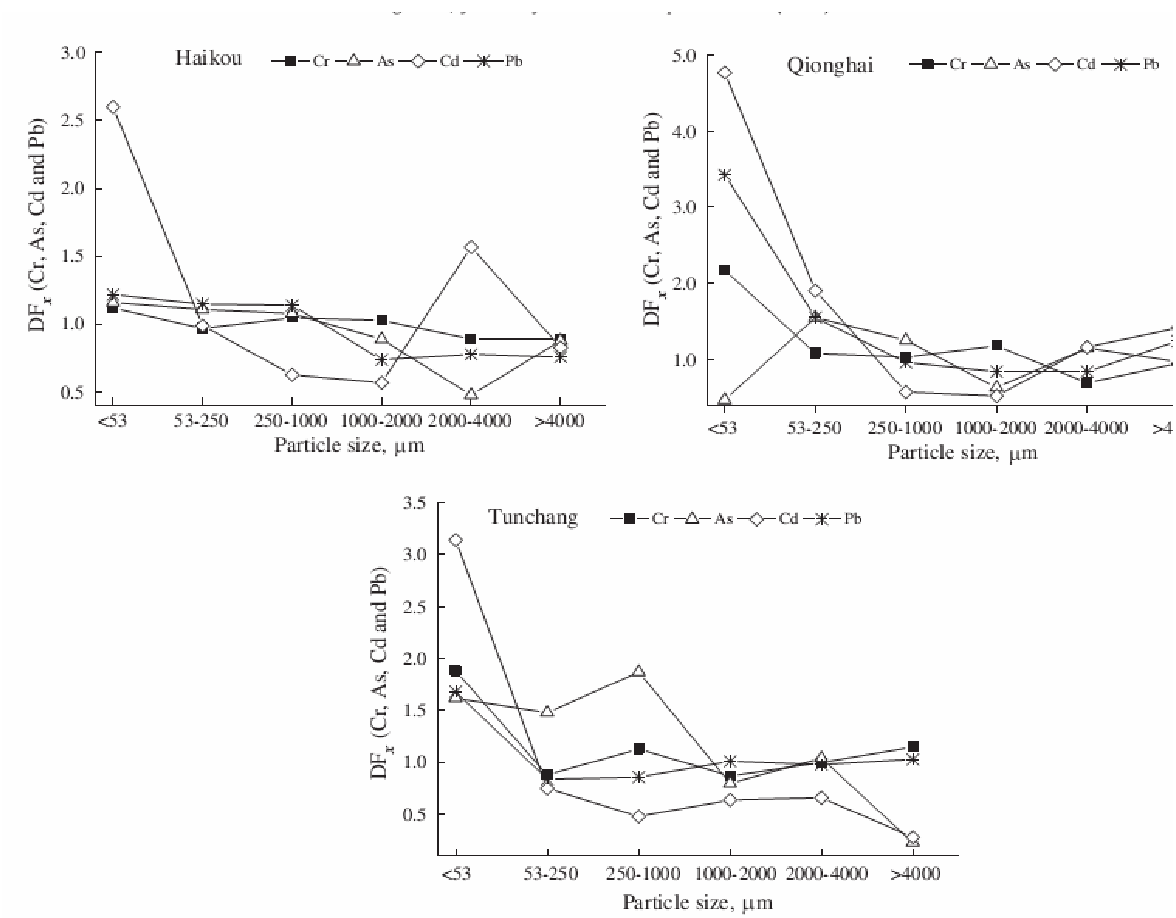


Fig. 3. Heavy metal mass loading (%) of six soil particle size fractions (<53 μm, 53–250 μm, 250–1000 μm, 1000–2000 μm, 2000–4000 μm and >4000 μm).

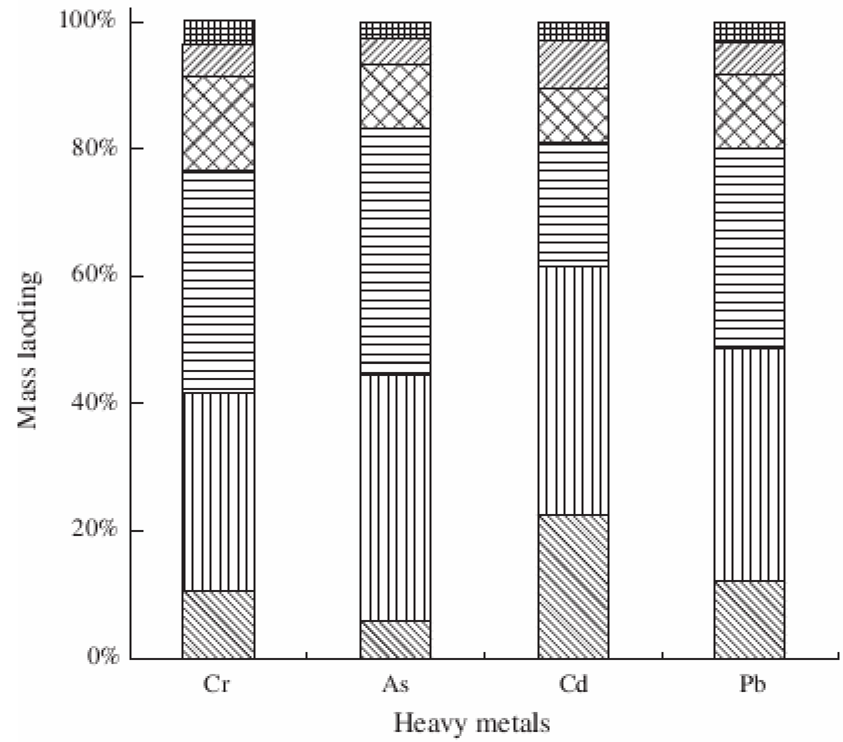


Fig.4 Leachability of heavy metals in different particle size fractions in arable soil

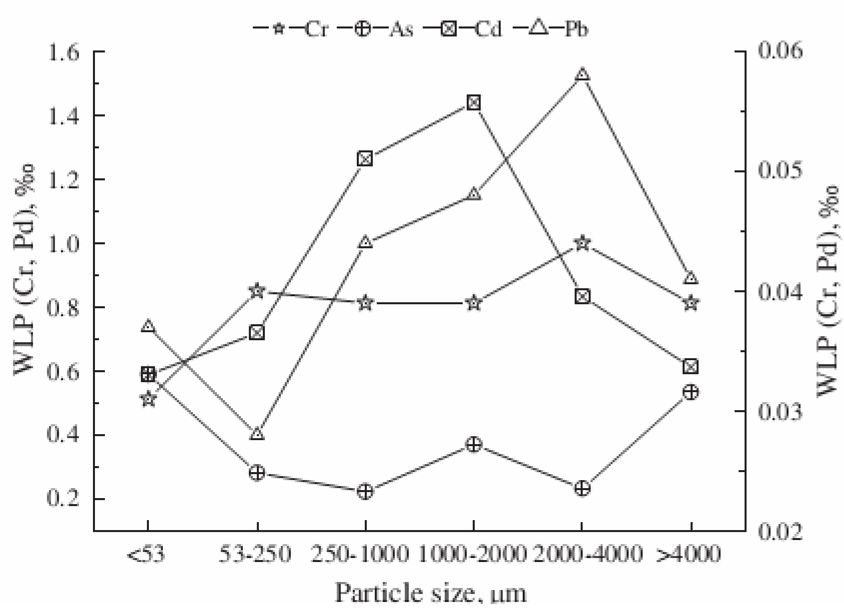
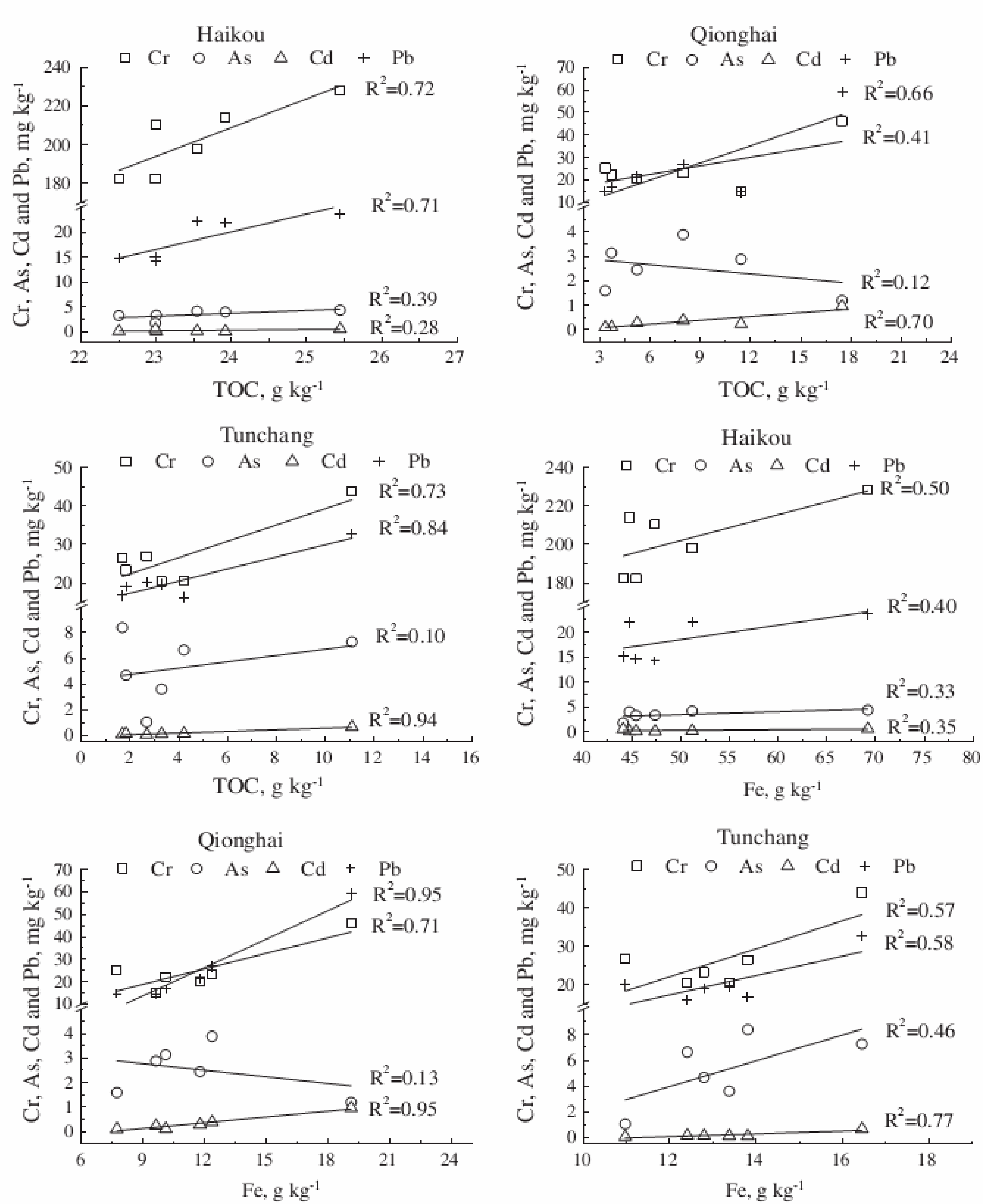


Fig.5 The correlations between heavy metals and the contents of organic carbons as well as Fe in different size fractions



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