

Environmental availability and profile characteristics of arsenic, cadmium, lead and zinc in metal-contaminated vegetable soils

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Abstract: Environmental availability and profile characteristics of arsenic (As), cadmium (Cd), lead (Pb) and zinc (Zn) were studied in contaminated vegetable soils from the Pb/Zn mining and smelting areas in Hunan Province of China, and the potential environmental risks of these metals were also assessed. The results show that the concentrations of As, Cd, Pb and Zn in vegetable soils are higher than the levels of Soil Environmental Quality of China (GB15618—1995). The mobility of metals in soil profiles is mainly characterized by the low pH and organic matter content of soil. The major part of As, Cd, Pb and Zn is restricted to the upper soils and the contamination of these metals in soils is significantly influenced by the long-term Pb/Zn mining and smelting activities. Based on the results from the BCR sequential extraction, the fraction of Cd in the soil profiles is predominantly existed in the acid-extractable form and the large amount of Pb is closely associated with reducible fraction. The environmental availability of Cd and Pb is predominantly higher than that of As and Zn in the soil profiles, suggesting Cd and Pb have more huge potential risk for human health and surrounding environment.

Key words: toxic element; environmental availability; contamination risk; soil profile characteristics; industrial contamination

1 Introduction

Metals and metalloids in soil, especially toxic elements, such as As, Cd, Pb and Zn, which are common pollutants in contaminated soil by nonferrous metals industrial activities, have the potential to seriously degrade eco-system function[1] and can pose a significant toxicological risk to most biological organisms. These substances can enter the body by direct inhalation or ingestion of contaminated soils, plant or water and may bio-accumulate in the body[2].

Active fractions of metals, inhaled or digested by animals and humans, have the ability to pass through the contaminated food-chain, resulting in a cumulated risk to human health and ecological safety[3]. During recent decades, the pollution of As, Cd, Pb and Zn in nonferrous metals mining and smelting areas has attracted a lot of attention, including their concentration, fraction distribution and impact on surrounding soil for safety use

and vegetable growth from the contaminated zone[4–5]. These results show that the environmental availability of toxic metals is not only decided by their abundance in soils, but also by their fraction distribution. Metal distribution and availability in soil profiles can easily describe their potential impacts of contamination[1, 6–7]. However, environmental availability and profile characteristics of metals in different types of metal-contaminated vegetable soils have not been fully discussed.

Hunan Province of China, with more than 100 years history for mining and smelting activities, has been consequentially evidenced widespread metals contamination at the surrounding environments[8–13]. The study of metals contamination in soil has typically focused on metal source, concentration and spatial distribution around heavily industrialized sites and contamination mainly affected by As, Cd, Cu, Pb and Zn[8, 10–11]. High levels of As, Cd and Pb in vegetables grown on contaminated soils has also been reported[5].

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Therefore, it is important to study the available fraction of metals and assess their ecological hazard in soils[14], particularly in agricultural soils sited near nonferrous metals areas[9]. In this work, the environmental availability and profile characteristics of As, Cd, Pb and Zn in contaminated vegetable soils from the metalliferous mining and smelting areas in Hunan Province of China and in four representative soil profiles from the corresponding sampling sites were discussed: (1) to determine environmental availability and profile characteristics of As, Cd, Pb and Zn in the soils; and (2) to assess the potential environmental risk of these metals by their available concentration and soil characteristics.

2 Experimental

2.1 Soil sampling and pre-treatment

Soils from the vicinity of metalliferous mining and smelting areas are heavily contaminated according to the previous investigation[5, 11]. In this work, the 45 vegetable surface soil (0–25 cm) samples from nonferrous metals mining, processing and smelting areas in Hunan Province of China, which were taken at distances of 1.5–2 km from pollution sources, were collected. The four types of soils were included, each representing a field scenario: soil contaminated by an abandoned smelter (SM-1), by the pollution source from a long-term smelter (SM-2), by smelting, mining and processing activities (SM-3) and by the contamination origin from mine tailings dump (SM-4), respectively. Four representative vegetable soil profile samples (three layers of 0–25 cm, 25–50 cm and 50–75 cm for each profile, respectively) were also collected from the four sampling areas: profile sample of SM-1 was collected in SM-1 area which is derived from quarternary red clay, of SM-2 was from slate and purple sandstone in SM-2 area, of SM-3 was from purple sandstone in SM-3 area and of SM-4 was from sandstone in SM-4 area, respectively.

All soils collected were air-dried and homogenized by sieving through a 2 mm screen. A portion of the sieved samples was then finely ground to pass through a 0.25 mm sieve. The most soil samples belong to acidic soils with low organic matter content (Table 1).

Table 1 Basic properties of surface soils

District	Soil pH ^a	Organic matter/% ^a
SM-1 (<i>n</i> =12)	5.05±0.43	3.03±0.04
SM-2 (<i>n</i> =18)	6.89±0.8	4.14±0.03
SM-3 (<i>n</i> =8)	4.64±0.35	5.14±0.02
SM-4 (<i>n</i> =7)	6.31±0.88	3.34±0.03

n is the number of analysed samples; and the data are presented in mean value and standard deviation.

2.2 Analytical methods

Soil pH was determined by a pH meter in a ratio of soil to de-ionized water being 1:2.5 (w/v). Soil organic matter was measured by the Walkley-Black procedure (consisting of oxidation of soil organic matter in 0.5 mol/L potassium dichromate and concentrated sulphuric acid with back titration of excess dichromate by standard ferrous sulphate)[15]. Particle size distribution was measured by hydrometer technique[16]. Operational fractions of metals in soil were determined through a three-stage BCR sequential extraction procedure as described by DAVIDSON et al[17]: 1.0 g dried soil was extracted for 16 h with 40 mL 0.11 mol/L acetic acid in a polypropylene centrifuged tube at 20 °C (step 1, acid-extractable fractions). The washed residue from step 1 was extracted for 16 h with 40 mL NH₂OH·HCl (0.5 mol/L, adjusted to pH 2.0 with nitric acid) at 20 °C (step 2, reducible fractions). The washed residue from step 2 was treated with 10 mL H₂O₂ (8.8 mol/L, adjusted to pH 2.0 with nitric acid) twice at 85 °C in a water bath for 1 h. The volume of liquid was reduced to about 1 mL and 50 mL of ammonium acetate (1.0 mol/L, adjusted to pH 2.0 with concentrated nitric acid) was added to the cooled moist residue and shaken for 16 h at 20 °C (step 3, oxidizable fractions). The total extractable fractions (*E*), including the acid-extractable fraction (*E*₁), reducible fraction (*E*₂) and oxidizable fraction (*E*₃), were calculated as the sum of these fractions, and were compared with the total concentration (*T*) of toxic metals in soils by digestion using a mixture of HF-HNO₃-HClO₄[15] in order to show the environmental availability of toxic elements. All of toxic elements in solutions were analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES, Intrepid II XSP, USA). Quality assurance of metal analysis was assessed using duplicates, method blanks and standard reference material for soil (GBW 08303).

2.3 Statistical analysis

The data were log-transformed prior to statistical analysis. All calculations were done with Excel 2003 and SPSS 11.0.

3 Results

3.1 Total and available concentrations of metals in surface soils

The vegetable soils are mainly contaminated by As, Cd, Pb and Zn. The concentrations of these metals in surface soils show great variability (Table 2). The geometric mean concentrations of As, Cd, Pb and Zn in soils substantially exceed the levels of Soil Environmental Quality of China (GB15618—1995)[18],

and the contamination of metals in the vegetable soils from the vicinity of metalliferous mining and smelting areas is heavily affected by the industrial activities. The available fraction concentrations for metal elements, including As, Cd, Pb and Zn, are also relatively high in surface soils. Most of Cd reveals as the most mobile major compound with high concentrations of extraction in E_1 (step 1). The concentrations in this phase varies from 0.21 mg/kg to 329.13 mg/kg (Table 2). The highest extractable amounts of Pb and As are found in E_2 (step 2) as expected. A part of Zn shows the huge environmental availability due to the high concentrations of acid-extractable and reducible fractions.

3.2 Concentrations distribution and availability for metals in soil profiles

Based on the studied soil profiles, the texture and organic matter contents in different soil profiles change significantly while the pH value of surface soils is mainly acidic and that of subsurface soils is mainly neutral (Table 3). The change of pH with sampling profile depth is important for controlling the mobility and profile distribution of metals. The organic matter content is the highest in surface horizons (0–25 cm) and greatly varies between surface and deep horizons (ranged from 5.15% to 0.65%) compared with the other soil layers (Table 3). The clay content ranges between 5%

and 47%, and the abundance of clay reaches the peak in the topsoils and declines at lower sampling depths.

3.2.1 Arsenic

The total As concentration in the whole soil profiles exceeds the levels of Soil Environmental Quality of China for agriculture (30 mg/kg, GB15618—1995) (Fig.1). Based on the distribution of total As concentration in the soil profiles at SM-1, SM-2 and SM-3, the majority of total As is restricted to surface soils (0–25 cm) while the total As concentration in the soil profile rapidly decreases with depth from 60 to 40 mg/kg at SM-1 and SM-3, and from 400 to 100 mg/kg at SM-2 (Fig.1). There is significant difference in transferring behaviour of As at SM-4 compared with the other sites. The total As concentration exceeds 1 000 mg/kg in the topsoil (0–25 cm) at SM-4, and is more than 400 mg/kg in the medium layer of soil (25–50 cm), while the high concentration of As (almost approaching to 1 000 mg/kg) is found in the bottom layer (50–75 cm). The result shows that there is seriously leaching of As in soil profiles at SM-4. But, the available fraction of As in all depths is very limited, and the role of available As in the profiles is negligible. The ratio between the total available fraction and total concentration ($E/T \times 100$) of As from the upper depth of 0–25 cm to the layer of 50–75 cm in soil is less than 10% (Table 3). The availability of As in the whole soil profiles is limited and

Table 2 Concentrations of total and extractable fractions of elements in soils ($n=45$)($\text{mg}\cdot\text{kg}^{-1}$)

Element	Total		E_1		E_2		E_3	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean
As	6.79–2 612.31	121.61	0.001–13.210	0.21	0.001–208.810	5.91	0.001–211.620	0.14
Cd	1.71–461.12	18.27	0.210–329.130	4.93	0.016–93.320	1.74	0.300–3.230	0.83
Pb	45.91–8 199.32	865.38	0.320–1 272.690	27.84	19.750–4 230.810	238.73	0.001–256.540	0.42
Zn	121.23–10 946.61	954.91	0.001–2 144.180	52.47	5.770–2 089.430	98.82	0.001–352.320	2.13

Table 3 Properties and ratio between total available fractions and total concentrations for As, Cd, Pb and Zn in contaminated soil profiles

Site	Depth/cm	pH (1:2.5, w/v)	Organic matter content/%	Clay content/%	Ratio/%			
					As	Cd	Pb	Zn
SM-1	0–25	5.50	3.06	22	3.72	83.30	21.06	19.86
	25–50	6.87	1.19	20	2.56	60.82	11.35	28.83
	50–75	6.84	2.85	19	2.20	74.58	16.20	27.36
SM-2	0–25	7.72	4.16	34	2.78	54.57	56.35	24.83
	25–50	7.43	3.68	24	6.57	50.94	58.69	22.62
	50–75	7.92	1.53	22	1.95	19.07	24.07	28.65
SM-3	0–25	4.65	3.43	47	3.02	48.16	48.18	23.28
	25–50	6.78	5.15	44	9.62	87.05	31.42	20.43
	50–75	7.25	4.39	40	0.68	48.72	84.42	25.16
SM-4	0–25	5.66	3.37	25	6.47	47.46	62.24	21.09
	25–50	6.64	2.11	17	3.57	35.17	49.13	22.79
	50–75	6.35	0.65	5	1.65	35.12	60.83	23.88

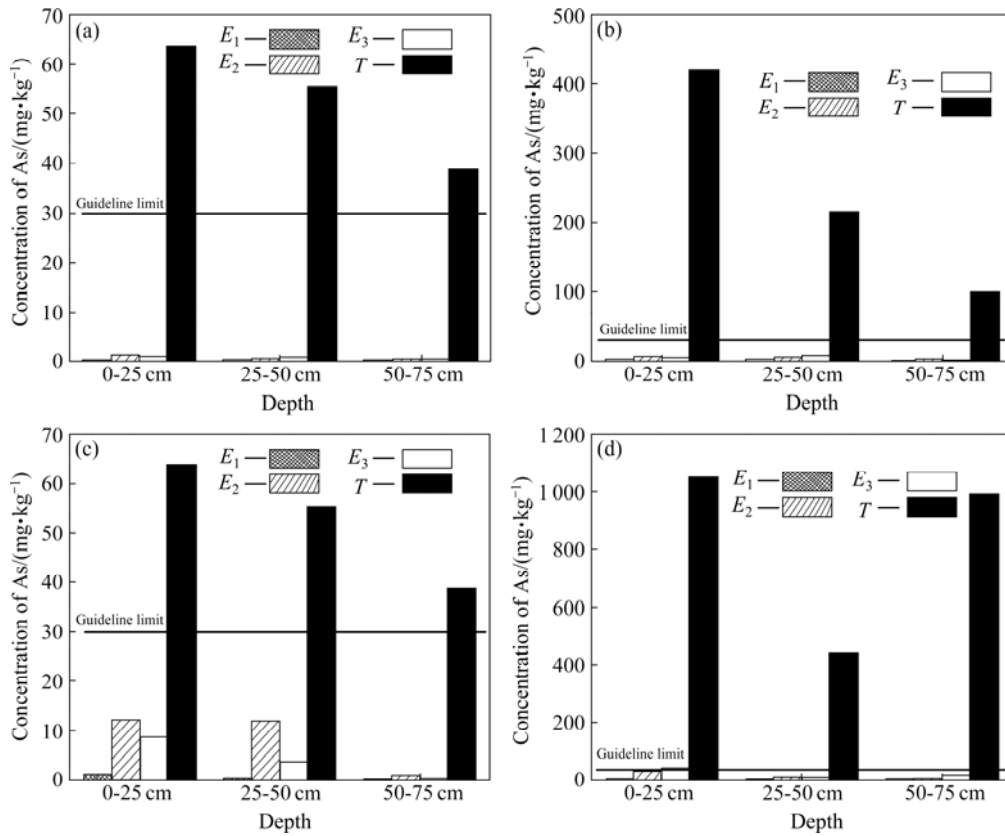


Fig.1 Concentrations of acid-extractable (E_1), reducible (E_2), oxidizable (E_3) and total (T) As in soil profiles: (a) SM-1; (b) SM-2; (c) SM-3; (d) SM-4

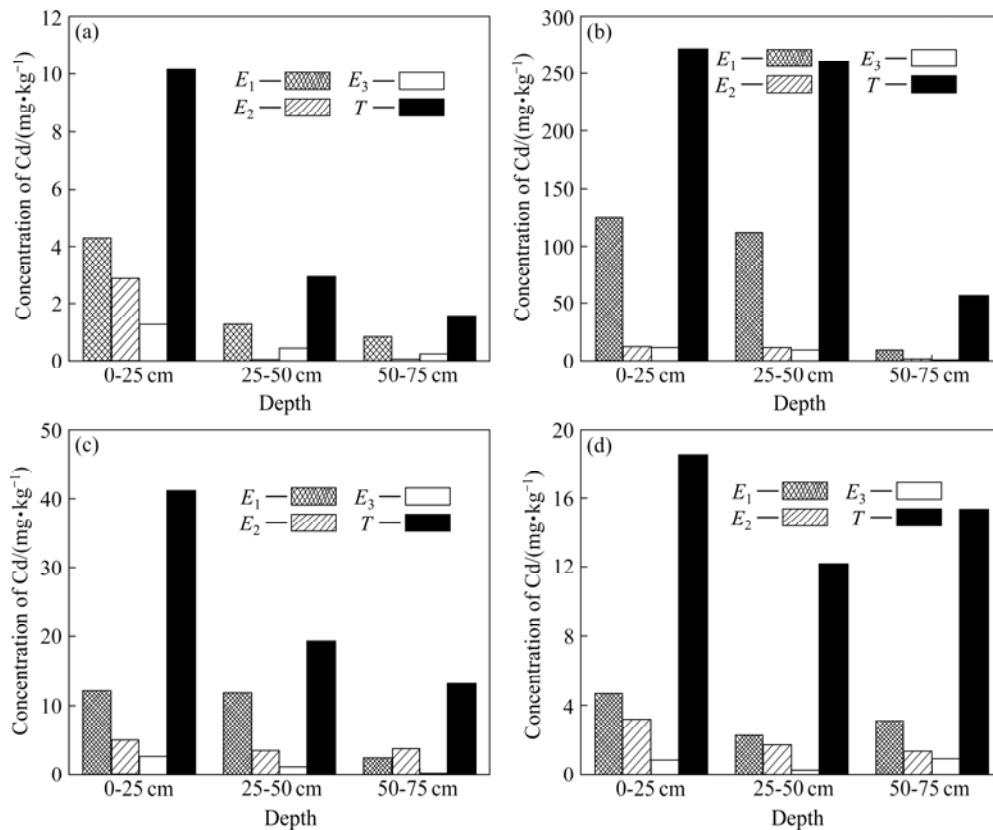


Fig.2 Concentrations of acid-extractable (E_1), reducible (E_2), oxidizable (E_3) and total (T) Cd in soil profiles: (a) SM-1; (b) SM-2; (c) SM-3; (d) SM-4

most of As is associated with the residual fraction in all investigated sites.

3.2.2 Cadmium

The cadmium concentration in soils ranges from 1.5 to 270 mg/kg, and is the highest in surface soils compared with the other layers (Fig.2). The concentration of acid-extractable Cd is easily transferred into the environment and exceeds 100 mg/kg at SM-2 with the sampling depths from 0–25 cm to 25–50 cm. It is significantly higher than that observed for the other three sites, where the concentrations of the fractions generally remain to be more or less than 10 mg/kg (Fig.2). In all soils, the concentrations of Cd fractions, in particular of acid-extractable fraction, are significantly higher than the levels of Soil Environmental Quality of China for agriculture (0.3 mg/kg, GB15618—1995). The ratio between the total available fraction concentration and total concentration of Cd ranges from 47.46% to 83.30% in the topsoil (0–25 cm), from 35.17% to 87.05% in the medium layer of 25–50 cm, and from 19.07% to 74.58% in the bottom soil (50–75 cm) (Table 3), respectively, indicating that the rate obtained for available Cd covers an appreciably large proportion of Cd in the whole soil and potential risk of Cd for human health and ecosystem function is extensive.

3.2.3 Lead

The concentration of Pb in the soil profiles is very high and decreases rapidly with the sampling depths at SM-1 and SM-3. The concentration of Pb decreases abruptly from 600 to 100 mg/kg at SM-1 and from 3 500 mg/kg to very low level at SM-3, respectively (Fig.3). The results show that the major part of Pb is restricted to the upper soils. On the other hand, the migration of Pb SM-2 and SM-4 is obviously different from the other sites. Pb concentration is higher in the 25–50 cm depth layer (more than 1 200 mg/kg) compared with the surface and lower layer at SM-2. The highest concentration of Pb at SM-2 is obtained in the deepest soil layer and arrives to 2 000 mg/kg. The extractable Pb in soil profiles mostly occurs in the reducible fraction. The ratio between the total available fraction concentration and total concentration of Pb in the whole soil profiles at SM-1 is very limited (Table 3). However, in the other sampling sites, the ratio of Pb ranges from 48.18% to 62.24% in the topsoil (0–25 cm), from 31.42% to 58.69% in the medium layer (25–50 cm), and from 24.07% to 84.42% in the bottom (50–75 cm), respectively (Table 3). It can be concluded that the environmental availability of Pb is high in these soil profiles and the potential risk is also significant.

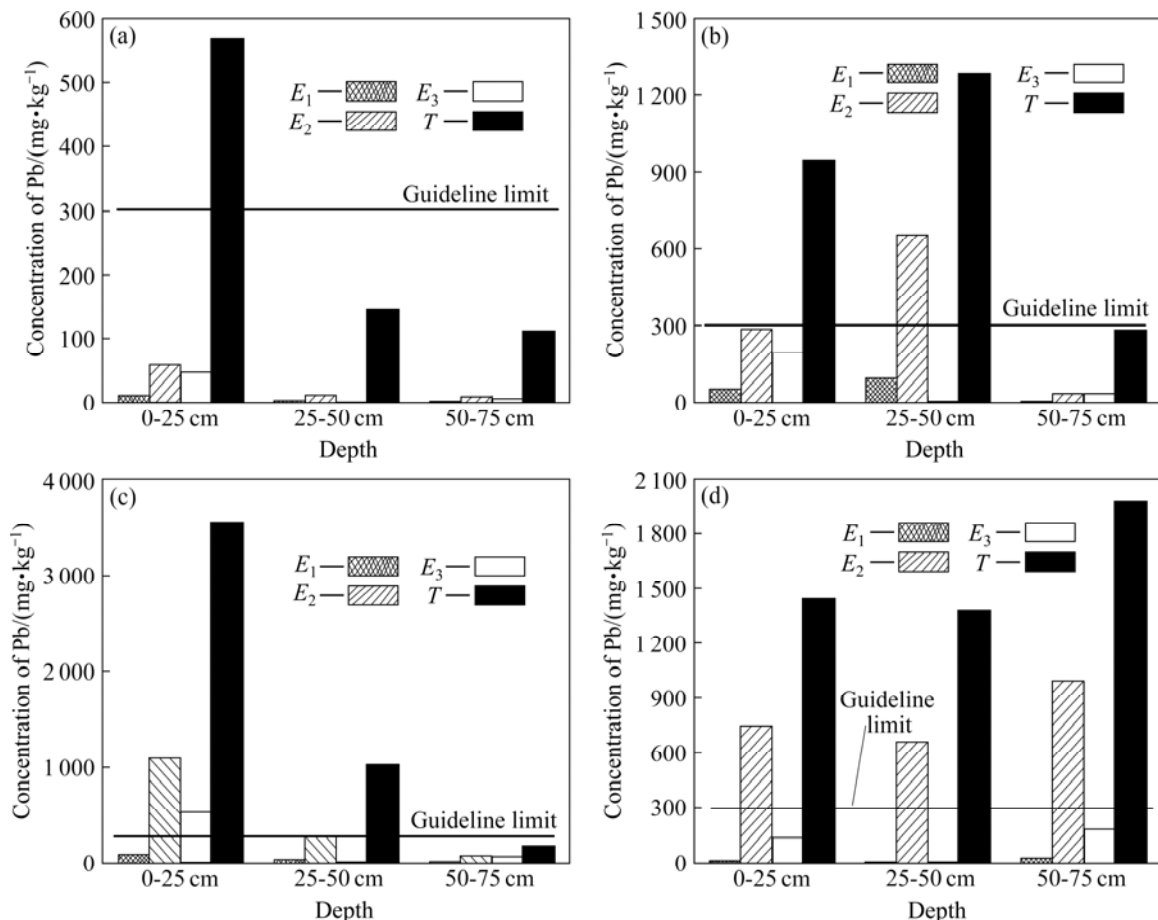


Fig.3 Concentrations of acid-extractable (E₁), reducible (E₂), oxidizable (E₃) and total (T) Pb in soil profiles: (a) SM-1; (b) SM-2; (c) SM-3; (d) SM-4

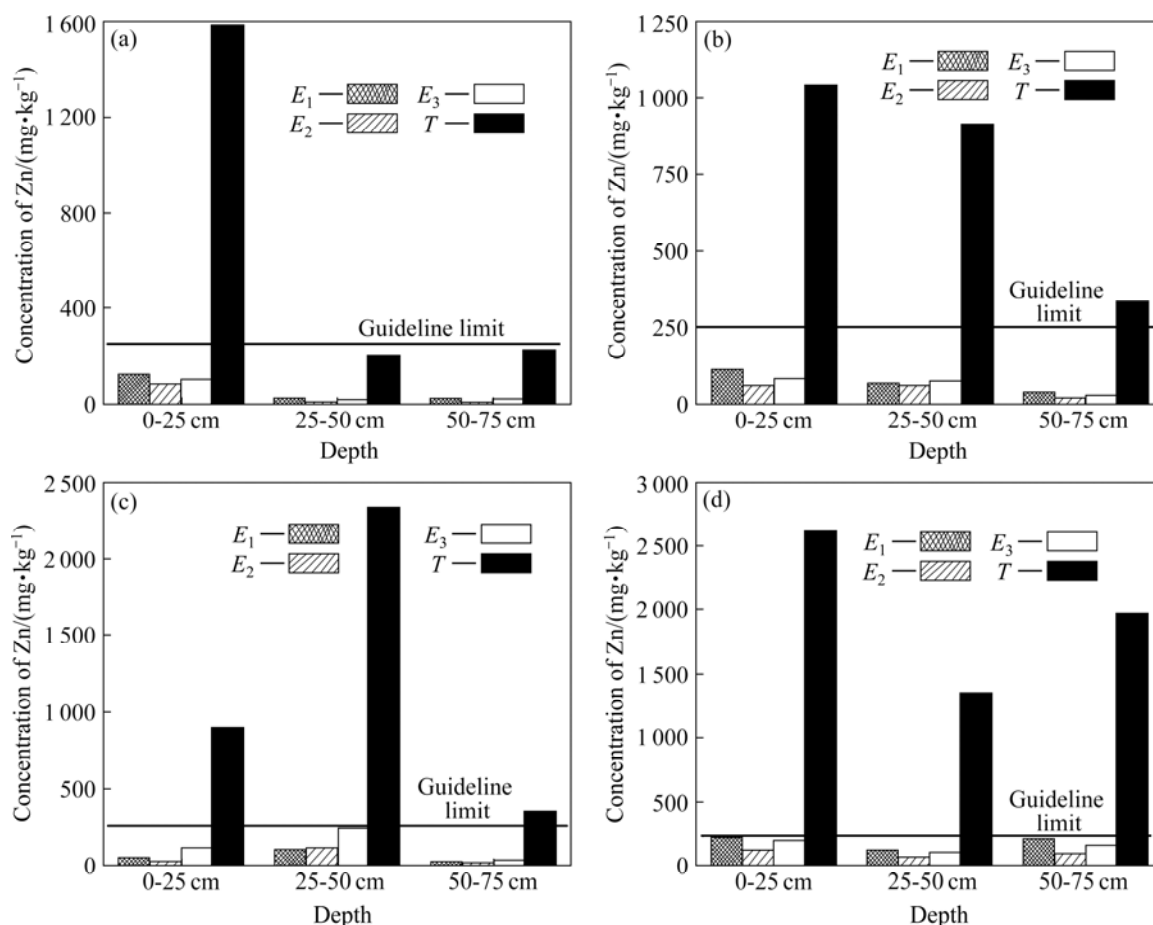


Fig.4 Concentrations of acid-extractable (E_1), reducible (E_2), oxidizable (E_3) and total (T) Zn in soil profiles: (a) SM-1; (b) SM-2; (c) SM-3; (d) SM-4

3.2.4 Zinc

Most of Zn accumulates in the upper soil of 0–25 cm and the concentration of Zn is 1 600 mg/kg at SM-1, 1 000 mg/kg at SM-2 and 2 500 mg/kg at SM-4, respectively. However, Zn has significantly migrated to the lower layer of 25–50 cm at SM-3 and the concentration of Zn arrives to 2 400 mg/kg (Fig.4). The result shows that the mobility of Zn in soils is significantly affected by soil properties. In most of soils, the total Zn concentrations at all depths exceed the levels of Soil Environmental Quality of China for agriculture (250 mg/kg, GB15618—1995) (Fig.4). But, available Zn is low and mostly associated with acid-extractable fraction. The ratio between the total available fraction concentration and total concentration of Zn in the whole soil profiles is almost between 20% and 30% (Table 3).

4 Discussion

In this study, the levels of toxic metals in metal-contaminated vegetable soils from the Pb/Zn mining and smelting areas, including As, Cd, Pb and Zn, are high and such a great amount of the four metals confirm a very serious polymetallic pollution. An accurate risk

assessment for heavy metals in contaminated soils should be considered both for the groundwater and for the aboveground vegetation[19]. The risk is closely dependent not only on metal concentration, but also on soil properties[19]. The studied acidic soil is characterized by low soil pH, high clay mineral and low organic matter contents (Table 3). The soil properties facilitate the environmental availability of heavy metals in the soil profiles.

The distribution of As, Cd, Pb and Zn in the contaminated vegetable soil profiles is significantly influenced by the contamination source. In this study, for Cd, the contaminant is deposited in large quantities in the all-sampling soils and seriously leached. The concentration of Cd in soil is significantly higher than the level of Soil Environmental Quality of China for agriculture (GB15618—1995) (Fig.2), which shows Cd contamination in the soils from mining and smelting areas is serious. For As, Pb and Zn, they are seriously contaminated (Figs.1, 3 and 4). However, the concentrations of As and Pb in the bottom layer of 50–75 cm at SM-4 are very high, which was contaminated by mining tailings containing a great deal of As and Pb after the mine tailing dam collapsed[9]. According to the

contamination of As, Cd, Pb and Zn in soil profiles at SM-2 and SM-3, the longer the historical/current contamination is, the stronger the leaching of metals in the soil profiles is. Besides, since it is known that the metals associated with total extractable fractions are more environmental available[20], the percentage of total available Cd in soil at SM-1 is higher than that of other samples, which shows that it is necessary to highly value the contamination of Cd in the historical contaminated soils, and the percentage of Pb in the current contaminated soils is also very high and the more attention should be paid to the potential risk for the local environment.

The extractable speciation of As, Cd, Pb and Zn in soil profiles can easily release into soil solution and surrounding environmental media. The acid-extractable As, reducible As and oxidizable As fractions represent a relatively small proportion of the total As in all soils, and demonstrate most of As is associated with the residual fraction in soils and very limited for environmental availability. In the study, the soil pH varies from acidic to neutral, and As mobility is quite low. Therefore, arsenic in soils would be more easily held in the surface soils. High concentration of Cd in acid-extractable fraction may lead to huge potential risk for environmental sustainability and human health. With a great amount of Cd in the acid-extractable fraction in the soils, it is clear that there is enough Cd for adsorption and accumulation from the soil matrix into plant tissues[21]. Lead is one of the least mobile metals in the soil, but Pb associated with Mn-and Fe-oxides could be mobilized into the environment as a result of these oxides dissolution. The fraction of Pb with high concentration is potentially accessible to the local environment. However, the concentration of Zn in soil profiles is very high while the acid-extractable fractions of Zn is very low, and it is slightly cared due to its necessity for vegetables in contaminated vegetable soils. On the other hand, the ratio between the total available fraction concentration by the three-stage BCR sequential extraction procedure and the total concentrations of Cd and Pb in the whole soil profiles are higher than those of As and Zn. The results show that the environmental availability of Cd and Pb is predominantly higher than that of As and Zn in the soil profiles, and Cd and Pb have more huge potential risk for human health and surrounding environment.

In all, it is essential for long-time natural remediation for vegetable soils heavily multi-contaminated by As, Cd, Pb and Zn. It is noteworthy that the soil pH and organic matter content are important for controlling the environmental availability of metals in soil profiles. So, the amendments, such as materials containing organic matter or additional pH-controlling measures (e.g. liming), should be used to change the

availability of metals in soil and greatly control the metals contamination in soil-vegetable system from the nonferrous metals mining and smelting areas.

5 Conclusions

1) The pollution in vegetable soils is mostly by As, Cd, Pb and Zn, and the environmental availability and risk of the toxic metals in the studied soil profiles is mainly characterized by soil pH and content of organic matter. The contamination of these metals in vegetable soils is also significantly influenced by the long-term effects of the Pb/Zn mining and smelting activities in the study areas.

2) The results from the BCR three-stage sequential extraction show that most of As and Zn in soil profiles are associated with the residual fraction; Cd is predominantly in the acid-extractable form, and the large amount of Pb is closely associated with reducible fractions. High concentration of acid-extractable Cd and reducible Pb in soil profiles could be easily transferred into the other environmental components, and may lead to huge potential risk for environmental sustainability and human health.

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