

Accumulation of heavy metals in soil and vegetables and absorption by field weeds with phytoremediation potential in fine chemical industrial park

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Received April 5, 2015

The Zhubugang area was a fine chemical industrial park area in Xiangtan city of China. Waste gas, water and slag discharges have caused serious heavy metals pollution to environment in this region. This study collected 10 soil samples, 5 varieties of seasonal vegetables (*Lactuca sativa*, *Lactuca sativa* L. var. *asparagine*, Bailey, *Radix Osterici Grosseserrati*, *Solanum tuberosum* and *Brassica rapachinensis*) and 3 varieties of field weeds (*Alopecurus aequalis Sobol*, *Alternanthera philoxeroides* (Mart.) Griseb, *Paspalum paspaloides* (Michx.) Scribn.). The concentrations of Zn, Cu, Pb and Mn were determined by hydride generation atomic fluorescence spectrometry after acid digestion. Then the environmental safety of heavy metals in soil and vegetables was evaluated and the absorption characteristics of field weeds to heavy metals were investigated. The results showed that the comprehensive pollution indices of heavy metals in the soils and vegetables were 0.79-3.43 and 1.54-4.13, respectively. Soil and vegetables were severely contaminated with heavy metals. The absorption capacity of different field weeds to heavy metals strongly differed. *Alternanthera philoxeroides* (Mart.) Griseb had the highest absorption capability to Cu of the 3 field weeds. The concentrations of Zn, Pb and Mn in *Alopecurus aequalis Sobol* were higher than in other weeds.

Key words: Zhubugang area; soil; vegetables; field weeds; heavy metals.

INTRODUCTION

The heavy metal pollution of soil becomes more and more serious in China in recent years. The heavy metal pollution has directly affected the health risk of residents. Chen Xing, Yang Gang, Zhang Xiaomin, Chen Hong and Wang Lixia et al. found that some lands in China such as arable land, mines, industrial area, city soil and river sediment were seriously contaminated with heavy metals [1-5]. According to the survey of the Ministry of agriculture, the soil of sewage irrigation was about $1.40 \times 10^6 \text{m}^2$, which means that almost 2/3 of the land area suffered from heavy metal pollution in China. The area ratio of slight pollution, middle pollution, serious pollution was 467:97:84 [6-8]. The agricultural land area contaminated with heavy metals was about $2.5 \times 10^7 \text{m}^2$. The food was contaminated with heavy metals at least $1.2 \times 10^{10} \text{kg/y}$. Released in 2014, "the national survey of Soil Pollution Bulletin" showed that the rate of soils with concentrations of pollution exceeding the standard was 16.1% of all soils of China. The soil pollution with industrial and mining waste was particularly prominent and the heavy metal pollution was the main pollution. The most

serious pollution in soil was cadmium of all heavy metal in China [9].

Heavy metals were extremely easy to be enriched in the soil. If not artificially processed, it may trigger social effects of pollution and serious harm to human, animals and plant life. Once the soil has suffered from heavy metal pollution, governance was difficult [10-14].

Zhubugang fine chemical zone area was located in Xiangtan City of Xiangjiang River Basin of China and was founded in 1960s. Xiangjiang River was the first river reported to be contaminated with heavy metals. The total area was $1.74 \times 10^6 \text{m}^2$. Heavy metal pollution was very serious in this area and Zhubugang area was the key heavy metal treatment area of Xiangjiang River Basin. The enterprises of this industrial zone mainly include chemical, smelting, electroplating and pharmaceutical enterprises. The volume of wastewater discharged was $2.64 \times 10^5 \text{kg}$ and contained $1.2 \times 10^6 \text{kg}$ of chemical oxygen, 218 kg of heavy metals, $6 \times 10^3 \text{kg}$ of sulfur dioxide and $3 \times 15 \text{kg}$ of industrial waste residue in 2013 by 26 enterprises in this area. Although pollution load significantly decreased in recent years through the implementation of government regulations, the land was still has not been able to grow because of the contamination with heavy metals [15].

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This study collected soil, vegetables and field weeds, analyzed Zn, Cu, Pb and Mn concentrations and evaluated the environmental safety of heavy metals in soil and vegetables, then investigated the absorption characteristics of field weeds to heavy metals. The conclusion of the study provided the scientific basis for the heavy metal treatment of soils in Zhubugang area.

MATERIALS AND METHODS

Collected samples

On April 20, 2014, we collected surface soil (0~20 cm) by the mixed sampling method where three samples were mixed into one sample in each point [16]. We collected plant samples in the soil sample point. The sampling location was shown in table 1.

Sample treatment

Plant sample

The edible parts of vegetables and the ground parts of field weeds were collected with a stainless steel knife. The dirt was first washed with tap water and rinsed 1 to 2 times with pure water, then washed 2-3 times with high purity water. Fixation was performed (105°C, 0.5 h) with a coarse filter paper wrap in the oven. Then the samples were dried to constant weight (80°C, about 12 – 24 h). The dried plant samples were milled in a mortar and finally put in a plastic bag for standby [16].

Soil sample

The sand and plant debris of soil were removed before natural air-drying. The soil was milled in a glass rod mill in a clean porcelain dish. Finally, the soil was filtered through 60 and 100 mesh nylon screens.

Sample analysis

At first, samples were placed in a polyethylene digestion tank and 6 ml 65% nitric acid and 2 ml 30% hydrofluoric acid were added. Then the lid was tightened and the vessel was placed in the microwave digestion instrument (USA CEM MARS 6) until the digestion tank wall dried after a period of time.

The concentrations of heavy metals in the samples were determined by flame atomic absorption spectrophotometry (Hitachi TAS 990F atomic absorption spectrophotometer) and graphite furnace atomic absorption spectrophotometry (Hitachi Z-2000 atomic absorption spectrophotometer) [16,17].

RESULTS AND ANALYSIS

Concentrations of heavy metals in soils and plants

Concentrations of heavy metals in soils

The concentrations of heavy metals in the soils were shown in table 1. Different Cu concentrations were found in the different soils, being higher in soils No.6 and No.2 and lower in soils No.9 and No.10. The concentrations of Cu in the soil didn't exceed the standard of soil environment quality. The concentrations of Mn in the soil differed significantly and the soil No.2 was of the highest Mn concentration. The concentrations of Pb exceeded the standard of soil environment quality in soils No.2, No.4 and No.6. The concentrations of Zn displayed the highest differences in all soils and the range of concentrations was 43 mg/kg-364 mg/kg. The No.2 soil was of the highest concentration of Zn.

Table.1. Sampling locations.

Soil sample	Latitude	Longitude	Distance to factory	Soil	Plant
No.1 soil	N27° 54'30"	E112° 58'17"	1m	Sludge of outfall	—
No.2 soil	N27° 54'32"	E112° 58'16"	5m	Sludge of outfall	—
No.3 soil	N27° 54'34"	E112° 58'15"	15m	Garden soil	<i>Lactuca sativa L. var.asparagina Bailey</i> <i>Lactuca sativa</i>
No.4 soil	N27° 54'23"	E112° 58'08"	25m	Paddy soil	—
No.5 soil	N27° 54'34"	E112° 58'16"	50m	Paddy soil	—
No.6 soil	N27° 54'46"	E112° 58'11"	200m	Garden soil	<i>Lactuca sativa L. var. asparagine Bailey</i> <i>Radix Osterici Grosseserrati</i>
No.7 soil	N27° 54'36"	E112° 58'18"	250m	Paddy soil	—
No.8 soil	N27° 54'38"	E112° 58'20"	400m	Paddy soil	<i>Alopecurus aequalis Sobol Alternant</i> <i>heraphiloxeroides (Mart.) Griseb Paspalum</i> <i>paspaloides (Michx.) Scribn.</i>
No.9 soil	N27° 55'08"	E112° 58'20"	1500m	Garden soil	<i>Solanum tuberosum</i>
No.10 soil	N27° 55'15"	E112° 58'23"	1800m	Garden soil	<i>Brassica rapachinensis</i>

There was no evaluation of the concentration of Mn in the soil environment quality of China. The background value of national soil was 583.0 mg/kg. The concentrations of Mn in soils No.1, No.2, No.3 and No.4 were higher than the background value of national soil.

The concentrations of heavy metals in vegetables were shown in table 3. As can be seen, the concentrations of heavy metals in the different vegetables were different. The vegetable with the highest concentrations of Cu and Mn was *Brassica rapachinensis*. The vegetable with the highest concentrations of Pb and Zn was *Lactuca sativa*. The concentrations of Pb in 3 vegetables were higher than the limit standard of this heavy meal of vegetable.

Table 2. The concentrations of heavy metals in soils (mg/kg).

Soil sample	Cu	Mn	Pb	Zn
No.1 soil	12.1b	604.4b	19.0b	74.3b
No.2 soil	56.4a	1060.2a	89.4a	360.5a
No.3 soil	44.3a	927.1a	76.3a	45.2b
No.4 soil	48.2a	768.1ab	88.7a	364.2a
No.5 soil	36.8ab	506.7b	58.5ab	324.4a
No.6 soil	62.4a	575.8b	74.4a	43.7b
No.7 soil	32.2ab	354.3c	73.2a	308.9a
No.8 soil	28.7 ab	248.5c	59.9ab	202.5a
No.9 soil	20.5b	322.9c	25.6b	49.3b
No.10 soil	20.0b	365.4c	27.5b	49.1b
Soil environment quality [18]	100	—	50/80	250
Background value of national soil [19]	22	583.0	26	74.2

There was a limit standard of Cu and Zn in vegetables in GB/T 5009.13—2003[21], GB/T 5009.14—2003[22], but the limit standard of Cu and Zn in vegetable was canceled in the new standard GB 2762—2012[19]. The main reason was the consideration that the concentrations of Cu and Zn in vegetables would not lead to problems of food safety in China. But the researchers believed that there was still risk in heavy metal polluted places. The related standard of the concentration of Mn in vegetables was not always made in China. But there was the general concentration of Mn in vegetables in "China food composition table (second volume)" [23] and the general concentrations were different in the various vegetables. We would use the data about Cu, Zn and Mn of GB/T 5009.13—2003, GB/T 5009.14—2003, and "China food composition table (second volume)" to evaluate the safety of vegetables. It was necessary to comprehensively

evaluate the food safety of every heavy metal in the vegetables because the amounts of eaten vegetables by the residents were grown locally.

Environmental safety of heavy metals in soils and vegetables

The pollution evolution method of heavy metals

The pollution evaluation method of heavy metals adopted in this study was the N. L. Nemerow index method. The latter was a comprehensive pollution evolution method. The method was presented with formula 1.

$$P = \sqrt{\frac{(Ci/Si)_{max}^2 + (Ci/Si)_{ave}^2}{2}} \quad (1)$$

Ci was the heavy metal concentration of sample; Si was the standard value of heavy metal; Ci/Si was the single pollution index No.i heavy metal; max was the maximum single pollution index; ave was the average single pollution index [24]. The five soil pollution grades according to the N. L. Nemerow index method [23] were: safe ($P \leq 0.7$), warning ($0.7 < P \leq 1$), slight ($1 < P \leq 2$), middle ($2 < P \leq 3$) and serious ($P > 3$).

The pollution evolution of soils

The pollution evolution results of soil were shown in table 4. Through comprehensive evaluation of Zhubugang soil, the soil heavy metal pollution was estimated as very serious in this region and the most serious pollution was that of Zn (comprehensive pollution index of 3.03). The Zn pollution of soils No.4, No.2, No.5, No.7 and No.8 was relatively serious in 10 soil samples, where the range of the single factor pollution index was 2.13 - 3.84. The comprehensive pollution index of Pb (2.54) was the second one among the 4 heavy metals. The pollution grade of Pb was middle. The concentrations of Pb in 7 soil samples exceeded the standard value according to the single pollution index. The pollution grade of Mn and Cu was slight, but 6 soils exceeded the standard values for Mn and 7 soils – that for Cu. The comprehensive pollution indexes of Mn and Cu were 1.86 and 1.68 respectively.

A comprehensive analysis of 10 soil samples, table 4, showed that the No.2 soil sample had the highest comprehensive pollution index of 3.43. Second was the No. 4 soil sample with comprehensive pollution index of 3.37, followed by No. 8 and No. 9 soils, which, having comprehensive pollution indices of 2.8554 and 2.76, were in the middle pollution grade. In the middle pollution grade were also the No.3 and No. 6 soils with comprehensive pollution indices of

2.17 and 2.02. No. 1 and No. 8 soils with comprehensive pollution indices between 1 and 2, were in the slight pollution grade. The control soils No.9 and No. 10 had comprehensive pollution indices of 0.77 and 0.83, thus being in the warning pollution grade.

Comprehensive tables 1 and 4 showed that the soil located in the sewage outfall of the same production factory waste, No. 1 soil was more seriously polluted than No. 2, and the difference in the comprehensive pollution index was about 3 times. Based on the sampling situation, No. 2 soil in a partial underground sewage ditch, and its surrounding still did not stop factory production, the wastewater might still be discharged into the soil around the factory. The No. 1 soil was located in an open sewage outfall, and the soil had dried after a long time of natural precipitation, low pollution soil around of the sewage outfall mixed with No. 1 soil, thereby reducing the heavy metal pollution.

We found the comprehensive pollution index of

No. 4 paddy soil (1.2) higher than that of No. 3 garden soil. At the same time, the comprehensive pollution index of No.7 paddy soil (0.7) was higher than that of No. 6 garden soil. Therefore, the heavy metal pollution of the paddy soil was more serious compared to the garden soil at the same distance to the factory in Zhubugang area. The main reason might be related to sewage irrigation in the farmland over a long period, because the wastewater of the factory in Zhubugang area was directly discharged into the nearby ditch used for farmland irrigation. We can see from table 3 that the degree of pollution of paddy soil and garden soil continuously decreased with the increase of the distance from the factory. The No.9 and No.10 garden soils had the smallest comprehensive pollution index among all soils, but their pollution grade was warning. This showed that there was a serious threat to the soil 1500 m from the factory. Measures must be taken to prevent the further spread of pollution.

Table 3. The concentrations of heavy metals in vegetables (mg/kg fresh weight).

Vegetable sample	Cu	Mn	Pb	Zn
<i>Lactuca sativa L. var. asparagine Bailey I</i>	0.40b	0.80c	0.20c	1.00b
<i>Lactuca sativa</i>	0.53ab	0.40c	1.06a	2.11a
<i>Lactuca sativa L.var. asparagine Bailey II</i>	0.60a	0.80c	0.20c	0.20c
<i>Radix Osterici Grosseserrati</i>	0.55ab	0.55c	0.37bc	2.03a
<i>Solanum tuberosum</i>	0.36b	1.98b	0.18c	0.18c
<i>Brassica rapachinensis</i>	0.62a	3.74a	0.62b	0.21c
The limit standard of heavy metal in vegetable [20]	—	—	0.3	—

Table 4. The pollution evolution results of soils.

Soil	Single pollution index				Comprehensive pollution index	Pollution grade
	Cu	Mn	Pb	Zn		
No.1 soil	0.44	1.32	0.64	0.78	1.13	Slight
No.2 soil	2.05	2.31	3.00	3.80	3.43	Serious
No.3 soil	1.61	2.02	2.56	0.48	2.17	Middle
No.4 soil	1.76	1.67	2.96	3.84	3.37	Serious
No.5 soil	1.32	1.10	1.95	3.41	2.86	Middle
No.6 soil	2.27	1.25	2.49	0.45	2.02	Middle
No.7 soil	1.17	0.77	2.46	3.25	2.76	Middle
No.8 soil	1.03	0.54	1.99	2.13	1.86	Slight
No.9 soil	0.73	0.70	0.84	0.52	0.77	Warning
No.10 soil	0.73	0.80	0.91	0.52	0.83	Warning
Comprehensive pollution index	1.85	1.68	2.54	3.03		
Pollution grade	Slight	Slight	Middle	Serious		

Table 5. The pollution evolution results of vegetables.

Vegetable	Single pollution index				Comprehen- -sive pollution index	Pollution grade
	Cu	Mn	Pb	Zn		
<i>Lactuca sativa</i> L. var. <i>asparagine</i> Bailey I	0.40	2.67	0.67	2.50	2.18	Middle
<i>Lactuca sativa</i>	0.53	0.67	3.53	5.28	4.13	Serious
<i>Lactuca sativa</i> L. var. <i>asparagine</i> Bailey II	0.60	2.67	0.67	0.50	2.04	Middle
<i>Apium graveolens</i>	0.55	1.38	1.23	5.08	3.87	Serious
<i>Solanum tuberosum</i>	0.36	1.98	0.90	0.45	1.54	Slight
<i>Brassica rapachinensis</i>	0.62	2.49	2.07	0.53	2.03	Middle
Comprehensive pollution index	0.68	2.35	2.50	3.73		
Pollution grade	Safe	Middle	Middle	Serious		

Table 6. The concentrations of heavy metals in field weeds (mg/kg dry weight).

Field weeds	Cu	Mn	Pb	Zn
<i>Alopecurus aequalis</i> Sobol.	8.0	952.3	53.4	106.7
<i>Alternant heraphiloxeroides</i> (Mart.) Griseb.	12.2	359.4	16.3	21.6
<i>Paspalum paspaloides</i> (Michx.) Scribn.	8.5	260.9	8.1	12.4

Table 7. The enrichment coefficients of 3 field weeds.

Field weeds	Cu	Mn	Pb	Zn
<i>Alopecurus aequalis</i> Sobol.	0.29	3.84	0.90	0.52
<i>Alternant heraphiloxeroides</i> (Mart.) Griseb.	0.43	1.45	0.27	0.11
<i>Paspalum paspaloides</i> (Michx.) Scribn.	0.29	1.05	0.14	0.06

The pollution evolution of vegetables

The pollution evolution results of vegetables were shown in table 5. The highest pollution was for *Lactuca sativa* in all vegetables and the comprehensive pollution index was 4.13. The *Lactuca sativa* samples were contaminated with Zn and single pollution factor was up to 5.28. *Lactuca sativa* L. var. *asparagine* Bailey of No.3 soil was mainly contaminated with Mn and Zn, the single pollution indices were 2.67 and 2.50 respectively, and the comprehensive index was 2.18, belonging to the middle pollution grade. *Lactuca sativa* L. var. *asparagine* Bailey of No.6 soil was mainly contaminated with Mn, the single pollution indices were 2.67, and the comprehensive index was 2.08, belonging to the middle pollution grade. *Apium graveolens* of No.6 soil was mainly contaminated with Zn, the comprehensive index was 3.87, belonging to the serious pollution grade. The pollution grades of *Lactuca sativa* and *Apium graveolens* were serious. The comprehensive pollution index of Zn was the highest among the heavy metals and that of Cu was the lowest. The concentration of Cu was safe in all vegetables and that of Zn was serious.

In summary, the vegetable samples were polluted with heavy metals, and the pollution level of vegetables continuously decreased with the increase of the distance from the factory. The same kind of vegetable of different regions had different pollution grades; the different kinds of vegetables of the same region had different pollution grades

with heavy metals. The main reason of the serious pollution was exhaust emission, sewage discharge and sewage irrigation by the factory in Zhubugang area.

The heavy metals in field weed

Concentrations of heavy metals in field weeds

The paddy field around the factory in Zhubugang area didn't cultivate crops for 10 years, but there were many field weeds grown. *Alopecurus aequalis* Sobol., *Alternant heraphiloxeroides* (Mart.) Griseb. and *Paspalum paspaloides* (Michx.) Scribn. were better grown than the other wild plants.

The concentrations of heavy metals in field weeds were shown in table 6. It can be seen that *Alopecurus aequalis* Sobol. had the highest concentrations of Mn, Pb and Zn, *Alternant heraphiloxeroides* (Mart.) Griseb. had the highest concentrations of Cu and *Paspalum paspaloides* (Michx.) Scribn. had the lowest concentrations of the 4 heavy metals in the 3 field weeds.

Absorption ability to heavy metal off-field weeds

A plant was considered hyperaccumulator if the concentrations of heavy metal (Pb, Zn, Cu and Mn) in the plant were more than 1×10^3 mg/kg, 1×10^4 mg/kg, 1×10^3 mg/kg and 1×10^4 mg/kg, respectively; or the concentration of heavy metal was 10 times higher than in the normal plant; or the enrichment coefficient and the transfer coefficient were greater than 1 [25]. The calculation formula of the enrichment coefficient of plant was as follows:

$$An = Cn / Ci^{[26]}(2)$$

An was the enrichment coefficient of plant; Cn was the concentration of heavy metal of plant; Ci was the concentration of heavy metal of the soil where plant grows. The enrichment coefficients of 3 field weeds in No.10 soil were shown in table 7.

According to table 7 three kinds of weeds in paddy field were in accordance with Mn hyperaccumulator characteristics, but also showed different absorption ability to the same heavy metals in different plant species [27]. The absorption ability to Cu of *Alternant heraphiloxeroides* (Mart.) Griseb. was the strongest in 3 field weeds, and the absorption abilities to Mn, Pb and Zn of *Alopecurus aequalis Sobol.* were higher than those of *Alopecurus aequalis Sobol.* and *Paspalum paspaloides* (Michx.) Scribn. For the same plant, the order of absorption ability of *Alopecurus aequalis Sobol.* was Mn>Pb>Zn>Cu; that of *Alternant heraphiloxeroides* (Mart.) Griseb. was Mn>Cu>Pb>Zn; and that of *Paspalum paspaloides* (Michx.) Scribn. was Mn>Cu>Pb>Zn.

The concentrations of Mn in the 3 field weeds were 10 times higher than those in normal plants such as the vegetables in this study under the same conditions. So we thought of the 3 field weeds as hyperaccumulators to Mn. The concentration of Zn in *Alopecurus aequalis Sobol.* was significantly higher than in the normal plant, but the field weeds weren't significant accumulators of other heavy metals. The related research found that *Alternant heraphiloxeroides* (Mart.) Griseb. and *Paspalum paspaloides* (Michx.) Scribn. were hyperaccumulators of Cu, Pb and Zn [28,29]. But in our study, we couldn't confirm these results. Combined contamination by heavy metals could influence the absorption of heavy metals by plants.

CONCLUSIONS

(1) In the survey area, the soil was seriously contaminated with heavy metals. Through the investigation of local grown vegetables, it was found the soil was not suitable for growing vegetables, the boundary being 1800 m from the industrial zone. Heavy metal pollution was very serious in Zhubugang area. Local residents should be transferred outside the scope of heavy metal pollution as soon as possible.

(2) The absorption ability to Mn of 3 field weeds met the characteristics of a hyper-accumulating plant. Therefore, 3 field weeds could be important plants for repairing heavy metal pollution damages in local soil.

Acknowledgement. The authors gratefully acknowledge the financial support Chinese Academy of Sciences and Education Department of Hunan Province, China through the Research Foundation of STS Program (Grant No. KFJ-EW-ST-014) and Education Department (Grant No. 13B027) to conduct this study.

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НАТРУПВАНЕ НА ТЕЖКИ МЕТАЛИ В ПОЧВАТА И ЗЕЛЕНЧУЦИТЕ И АБСОРБЦИЯТА ИМ С ПОЛСКИ ПЛЕВЕЛИ С ФИТО-РЕМЕДИАЦИОНЕН ПОТЕНЦИАЛ

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².Ключова лаборатория по екологично ремедиация от тежки метали и безопасно използване на замърсени почви, Колеж на провинция Хунан, КсиангтанХунан, Китай

Постъпила на 5 април, 2015 г.

(Резюме)

Районът Жубуганг е бил индустриален парк за фина химическа промишленост в гр. Ксиангтан в Китай. Изхвърляните отпадъчни газове, води и шлаки са предизвикали сериозно замърсяване на околната среда с тежки метали. Това изследване се основава на 10 почвени проби, 5 различни сезонни зеленчуци (*Lactuca sativa*, *Lactuca sativa L.var. asparagine*, *Bailey*, *Radix Osterici Grosseserrati*, *Solanum tuberosum* and *Brassica rapachinensis*) и три вида полски плевели – *Alopecurus aequalis Sobol.*, (*Alternanthera philoxeroides (Mart.) Griseb.*, *Paspalum paspaloides (Michx.) Scribn.* Концентрациите на Zn, Cu, Pb и Mn в тях са определяни чрез атомна флуоресцент на спектрометрия след киселинно разтваряне и образуване на хидриди. Така са определени екологичната безопасност на тежките метали в почвата и абсорбционната способност на плевелите спрямо металите. Резултатите показват, че пълните индекси на замърсяване на почвата с тежки метали са съответно 0.79-3.43 и 1.54-4.13. Почвата и отглежданите зеленчуци са сериозно замърсени с тежки метали. Изследваните плевел има различна абсорбционна способност. Растението *Alternanthera philoxeroides (Mart.) Griseb* има най-висок абсорбционен капацитет спрямо медта от трите изследвани вида. Концентрациите на Zn, Pb и Mn в *Alopecurus aequalis Sobol* са най-високи спрямо останалите видове.