JOURNAL OF PURE AND APPLIED MICROBIOLOGY, May 2014. Vol. **8**(Spl. Edn. 1), p. 659-665

Cadmium Contents in Greenhouse Soils and Influencing Factors in Eastern Shandong Province, China

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(Received: 12 April 2014; accepted: 09 May 2014)

To understand the status of cadmium (Cd) accumulation and analyze the affecting factors in greenhouse soil under the intensive usage, 37 soil samples from greenhouse fields and 6 soil samples from local open fields were collected in eastern Shandong province. The range and average of Cd concentration in topsoil (0-20 cm) were 0.20-1.67 and 0.52 mg/kg, and 0.21-1.44, 0.47 mg/kg in subsoil (20-40 cm), respectively. 78.4 percent of the samples topsoil and 70.3 percent of subsoil exceeded soil environmental quality standard of China. It showed that Cd contamination of greenhouse soils in eastern Shandong is quite serious. Cd content in topsoil was extremely significant positive correlated with soil organic matter and total N. Due to the universal high input of organic/chemical fertilizers in the study area, Cd content in soil increased significantly with the increase of cultivating years. The present study indicates that long-term and excessive usage of nitrogen and organic fertilizer are an important cause leading to Cd accumulation in greenhouse field soils in eastern Shandong province. Most importantly, the widespread secondary salinization and acidification in greenhouse soil may further increase of Cd bioavailability in soil and accordingly enhance the health risk of people via eating vegetables.

Key words: Cultivating years, Facility vegetable soil, Heavy metal, Secondary salinization.

Cadmium (Cd), a persistent and toxic heavy metal, can result in many adverse health effects in a variety of tissues and organs. Cd intake by the human body through the contaminated food chain was the important exposure approach for the non-professional population[1,2]. Accordingly, reducing the cadmium uptake by roots has been one of the focuses of environmental science. The main source of soil Cd pollution was mining, smelting, electroplating and basic chemical industry wastewater, waste gas and waste residue[3,4]. Moreover, usage of chemical fertilizers, pesticides and agricultural sludge containing Cd was also another important source of soil Cd pollution.

In China, protected vegetable cultivation was developing rapidly with an area of 7000 hm^2 in 1982 and 4.67 million hm^2 in 2010[5]. By building structure facilities, the high yields of vegetable production could be achieved under artificial environmental condition[6]. However, the high strength usage of facility soil has brought a series of environmental problems such as secondary salinization, acidification and nutrient accumulation[7-9]. A large number of applications of chemical fertilizer and organic materials also resulted in Cd accumulation in greenhouse soil. According to the statistical analysis based on data from previous literatures published since 1989, about 24.1 percent of total samples were contaminated by Cd in Chinese vegetable soils[10]. In recent years, many investigations were conducted on heavy metals pollution in greenhouse soils in Shouguang city of Shandong province, Jilin, Liaoning and Jiangsu province[11-15].

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Results showed that Cd accumulation in vegetable field was higher than other heavy metals. The range of the average soil Cd contents from the different regions were 0.37-0.68 mg/kg, generally higher than the level in corresponding local open field soil, and the highest value was 5.2 times of that in open soil. Inevitably, Cd enrichment in soil could impose threat to Cd concentration in vegetables, further inducing potential adverse health impact.

Despite many documents concerning Cd pollution in greenhouse soils, few studies based on sampling and analysis in regional scale, reported the relationship between Cd contents and soil properties. The objective of the present work is to investigate the status of Cd contamination in greenhouse soils and analyze the relationship between Cd contents and soil physical and chemical properties and planting years by sampling from greenhouse soils in eastern Shandong province, an important facility agricultural area. It could provide a scientific basis for the prevention and control of Cd pollution in soil and safety production of greenhouse vegetables.

MATERIALS AND METHODS

Study area

Eastern Shandong province is located in 120°-122°43'E and 36°-38°24'N, adjacent to the Yellow Sea and on the south of Bohai Bay, including Weifang, Qingdao, Yantai and Weihai city. The area has typically temperate and monsoonal climate and four clearly distinct seasons, with average annual temperature 12.0-12.6°C and 650-850 mm of average annual precipitation. Loam is the major soil type, suitable for planting agriculture. As a developed area with a large population and relatively little arable land, facility vegetable cultivation has been developing rapidly and become the important base of facility vegetables in China, with planting history of three decades.

Sampling procedures

Sampling procedures were conducted from August to November in 2012. 37 samples from greenhouse field soil and 6 samples from local open field soil were collected from Weifang, Shouguang, Changyi, Gaomi, Qingdao, Pingdu, Jimo, Laixi, Laiyang, Haiyang, Zhaoyuan, Rushan, and Wendeng cities, which are located in eastern Shandong province, an important vegetable production base in China (Fig. 1). In the study area, the greenhouse facilities are not uniformly distributed. Accordingly, sampling sites are set in relatively concentrated areas of greenhouses in each city, meanwhile considering the soil type, planting years and fertilization etc. The major species of vegetables in the sampling area are tomatoes, cucumber, spinach, squash, beans, lettuce, garden chrysanthemum, towel gourd and ginger. At each spot, 6-10 subsamples were collected and mixed to one soil sample, and the soil sample was divided into top soil (0-20 cm) and subsoil (20-40 cm), in terms of the soil depth. Soil samples were air-dried, ground and sieved through 2 mm mesh for determining physical and chemical properties. The plastic or wooden tools were used in the sampling process to avoid pollution of metals. A part of each sample was ground and passed through a 0.149 mm nylon sieve for analysis of total Cd concentration. When the soil sample was collected, the relevant information such as cultivating years, fertilizing type and amount was enquired.

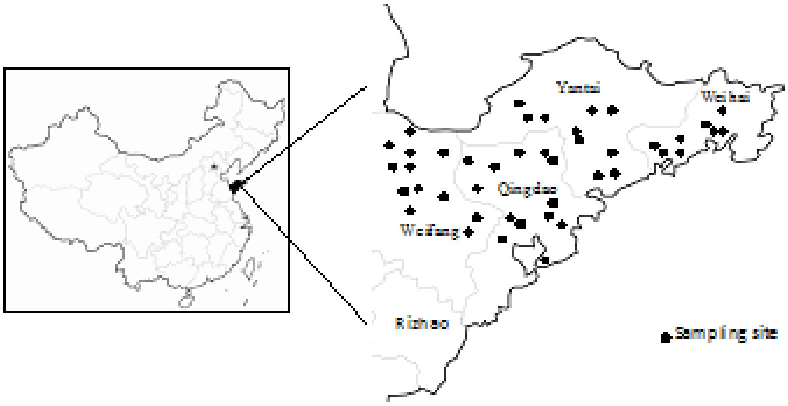

Fig. 1. Location sketch of sampling sites

Sample analysis and quality control

Soil chemical properties were measured using the method recommended by Lu[16]. Organic matter (OM) was determined with $K_2Cr_2O_7$ oxidation at 180ºC. Soil pH was measured in a 1:2.5 soil/water suspension with a combination electrode. Total N was determined with $K_2Cr_2O_7$-H_2SO_4 oxidation, total P was digested using a mixture of $HClO_4$-H_2SO_4. After digested in mixture of HNO_3-HF-$HClO_4$, Cd concentrations of soils were determined by AAS (AA-7000 Shimadzu Japan).

Glass apparatus used for sample analysis were immersed in 15% nitric acid (v/v) for 24 h, then thoroughly washed with deionized water before use. Cd contents were calculated via the standard curve method and reagent blanks were used in the analytical process. Certified standard soil samples GBW07401 (GSS-1) was used to ensure precision of the measurement. The recovery of the method was in the range of 86.4%–109.7%. Statistical analysis of the data was finished by using the SPSS software package (SPSS 17.0).

RESULTS AND DISCUSSION

Distribution and evaluation of Cd in soils

Cd concentration and distribution of topsoil (0-20 cm) and subsoil (20-40 cm) were listed in table 1. Soil cadmium content in two layer soil showed varied distribution status, normal distribution for topsoil and logarithmic normal distribution for subsoil. Range, average, geometric mean and mid-value were 0.20-1.67, 0.52, 0.48, 0.46 mg/kg in topsoil, and 0.21-1.44, 0.47, 0.42, 0.34 mg/kg in subsoil, respectively. Many studies have reported that heavy metal content increased with the increase of soil depth due to the agronomic measures resulting in accumulation of heavy metals in top soil[14, 17]. In this study, however, no significant variation of Cd content was observed between in topsoil and in subsoil ($p>0.05$), although topsoil presented a higher mean value. Furthermore, there was an extremely significantly positive correlation of Cd content between top soil and subsoil ($p<0.001$). It may be caused by disturbance of plant roots and frequent plough by human being in 0-40 cm soil. The high mobility of Cd in soil may also contribute to the consistence of the two layer soils in Cd content.

Table 1. Statistical analysis of Cd content in greenhouse soils (mg/kg)

Soil type	Maximum	Minimum	Average	Geomean	Median	Standard deviation
Topsoil	1.67	0.20	0.52	0.48	0.46	0.27
Subsoil	1.44	0.21	0.47	0.42	0.34	0.29

Compared with soil background value of Cd content in the study area (0.084 mg/kg)[18], all of greenhouse soil samples and almost all of open soil samples were contaminated by Cd due to human activities. This study area belongs to both vegetable farms and the greenhouse vegetable base. Soil environmental quality evaluation should be performed in term of China Environmental Quality Standard for Soils (GB 15618-1995) and China Environmental Quality Evaluation Standards for Farmland of Greenhouse Vegetables Production (HJ/T 333-2006). As a more mobile metal, cadmium content standard in soil was strictly controlled. For two standards, soil Cd content is limited 0.3 mg/kg in soils (pH<7.5). Referring to the standard, the method of soil single pollution index was used to evaluate the soil Cd pollution. Results showed that in 37 top soil samples, there were 29 samples exceeded the criterion with the over standard rate 78.4%. The maximum value reached 1.67 mg/kg, which is 5.6 times of standard limit. In subsoil samples, the over standard rate also reached 70.3% (Table 2). It indicated that Cd contamination in greenhouse soils in eastern Shandong is quite serious. Cd in soil is easier to transfer to vegetables especially for the root and leaf vegetables[19]. The aggravation of soil cadmium pollution may increase the risk of cadmium pollution of vegetables, which would pose a potential threat to human health.

Relationship between Cd content and soil properties

Soil properties are not only the embodiment of human activities on the soil for a long time, also influence the bioavailability of heavy

Table 2. Analytical table of exceeding standard of soil Cd contents

Type of sites	Number of sites	Layer type	Exceeding number	Exceeding rate	Maximal excess multiple
Open field soil	6	Topsoil	1	8.2%	0.3
		Subsoil	0	0	0
Greenhouse soil	37	Topsoil	29	78.4%	4.6
		Subsoil	26	70.3%	3.8

metals in the soil. The index of soil samples properties in the study area including pH, organic matter, total N and total P were presented in table 3. The range of pH in greenhouse soil was 5.51-7.62 and was average 6.61, showing an acidification trend compared with local open field soil, which was average pH 6.83. The consensus view currently accepted was that long-term and excessive usage of chemical fertilizers induced the accumulation of acid ions such as SO_4^{2-}0Cl^- and the relative decline of Ca^{2+}, Na^+, Mg^{2+} proportion, resulting in the reduction of soil pH value[8, 9, 20]. Under the high input of organic and chemical fertilizers, the contents of soil organic matter and total N were enhanced 1.4 and 0.74 times compared with open field soil, whereas the content of P showed a slight increase. This trend is largely consistent with fertilization habit of farmers who apply a lot of nitrogen and ignore other fertilizers[7]. No significant correlation was observed between Cd content and soil pH ($p>0.05$). However, as the most important factor affecting Cd mobility in soil, the aggravation of soil acidification could promote the bioavailability of Cd in soil via dissolution, ion exchange and desorption, accordingly promoting Cd uptake by plant roots[21-23]. Moreover, in the case of excess Cd accumulation in soil, the increase of the proportion of available Cd in greenhouse soil can further enhance the risk of Cd intake by people via eating vegetables.

Table 3. Physical and chemical properties of soil samples in study area

Soil properties	Range		Average		Standard deviation	
	Open Field	Green House	Open field	Green House	Open field	Green House
pH	5.88-7.68	5.51-7.62	6.83	6.61	0.08	0.09
OM (g/kg)	5.40-21.03	12.88-39.69	11.22	26.75	1.13	1.06
Total N (g/kg)	0.23-1.02	0.41-1.56	0.53	0.92	0.06	0.04
Total P (g/kg)	0.05-1.77	0.07-1.76	0.49	0.55	0.07	0.06

Cd content in topsoil increased with the increase of the content of organic material in soil and had a significant positive correlation ($P<0.001$) (Fig. 2). The increase of organic content in greenhouse soil was due to the application of a large number of organic materials, such as green manure, sewage sludge, and excrements of livestock. According to the surveying data from northern China, Cd content of pig manure from intensive feeding ranged from 0.22 to 2.93 mg/kg[24]. Apparently, a great quantity of organic fertilizers was the important factor inducing the accumulation of Cd in greenhouse soil. In addition, long-term use of organic fertilizer resulted in the increase of Cd uptake by crops due to the existence of some heavy metals[25-27]. The organic materials could reduce the bioavailability of Cd in soil by increasing Cd adsorption in soil. But on the other hand, the soluble organic matter such as organic acid could increase the mobility of Cd[28, 29]. The function of organic matter is related to the type of organic fertilizer, and also affected by environmental factors and soil properties. More attention should be given to the influence of increasing organic matter on the bioavailability of Cd in greenhouse soil.

To obtain high productivity, a large number of fertilizers are usually applied to the greenhouse soil by farmers. In Shouguang City, a famous greenhouse vegetable base, for example, the amount of fertilizer applied to greenhouse soil was as much as 3000-4500 kg/hm^2 for half a year and the residual amount of fertilizer in soil reached 90% in some area[7]. According to the survey, nitrogen fertilizer had the most amount of application in different chemical fertilizers, leading to large residues in soil. The result of correlation analysis presented a significantly positive correlation between Cd content and total N level in greenhouse soil ($p<0.01$) (Fig. 3). It indicated that in spite of low Cd content in N fertilizer, applying excessive amount of N fertilizer into the soil in a long term could also result in the accumulation of Cd in the greenhouse soil. Furthermore, various types of nitrogen fertilizer would aggravate soil acidification and ion accumulation in topsoil such as Cl^- and SO_4^{2-}, which has been confirmed to enhance Cd bioavailability in soil by the formation of complexes $CdCl^+$, $CdCl_2^0$ or $CdSO_4$[30-32].

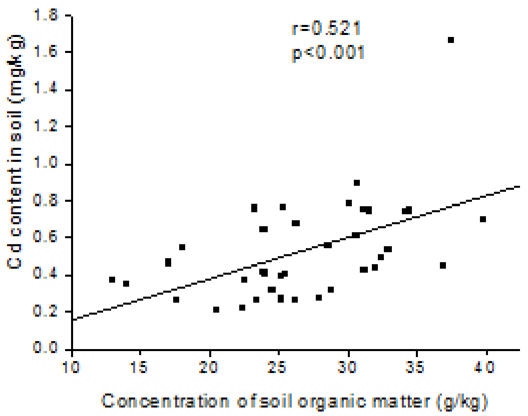

Fig. 2. Relationship between Cd contents of top soil and soil organic matter

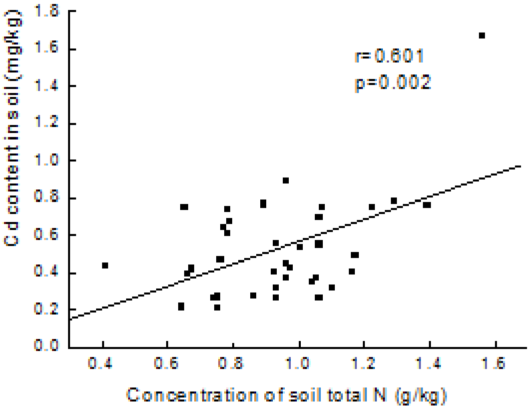

Fig. 3. Relationship between Cd contents of top soil and soil total N

Generally, due to more Cd in phosphate (P) fertilizer, the usage of P fertilizer could increase Cd content in soil and Cd uptake by plants[33, 34]. However, there was no significant correlation between Cd content and total P level in the greenhouse soil ($p>0.05$), which may be related to the less use of P and nutrient element fertilizers compared with N fertilizer and organic material in Shandong province. In addition, P fertilizer made in China has lower Cd content than imported fertilizers, which may partly account for the low accumulation of Cd induced by P application. On the other hand, phosphate can reduce the bioavailability of Cd in soil via formation of insoluble compounds as $Cd_{10}(PO_4)_6(OH)_2$[35]. In the soil of Cd contamination, the relative lack of P nutrition may further increase the risk of Cd adsorption by plant roots.

Effect of cultivating years on soil Cd content

Correlation analysis showed that there was no significant relationship between planting years and soil pH value, however, greenhouse soil showed acidification trend with the increase of planting years ($r=-0.024$). Soil organic matter and total N presented an extremely increasing trend with the increase of planting years ($p<0.01$). In our questionnaire survey in the study area, approximately 120-800 kg/mu chemical fertilizers were applied to soil as nitrogen or compound chemical fertilizers every year, which was 3-22 times of the amount in corn/wheat field soil. There was average 1400 kg/mu organic matter applied to soil as hog or chicken manure every year, whereas almost no manure applied to the corn field. The long term and excessive input are bound to result in the accumulation of heavy metals in greenhouse soil.

Compared with open field soils, Cd content in greenhouse soil increased in various degrees (Fig. 4). For topsoil, Cd content presented the increasing trend year by year with increasing cultivating years and reached the peak 0.82 mg/kg in the past decade, which was nearly twice of control, whereas a higher Cd content was found in

4-6 cultivating years for subsoil. Correlation analysis showed that Cd content in topsoil ($p<0.001$) and subsoil ($p=0.005$) were in extremely significantly positive correlation with cultivating years, which is in accordance with other reports[12, 36]. Heavy metals in greenhouse soil are originated mainly from organic manure, chemical fertilizer, pesticide and so on. However, due to the weak leaching mobility in soil and the limited amount taken away by plant, Cd could accumulate in top soil.

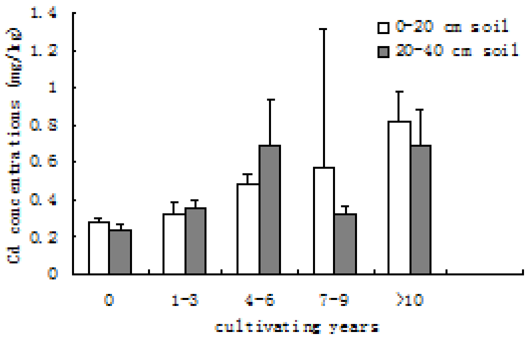

Fig. 4. Cd concentrations in greenhouse soils under different cultivating years

CONCLUSIONS

In greenhouse soil of eastern Shandong, the range and average of Cd concentration were 0.20-1.67 and 0.52 mg/kg in top soil (0-20 cm), and 0.21-1.44 and 0.47 mg/kg in subsoil (20-40 cm), respectively. Of the Cd contents in 37 soil samples, 78.4 percent of the topsoil and 70.3 percent of subsoil exceeded soil environmental quality standard of China. There is quite serious Cd contamination of greenhouse soils in eastern Shandong province.

Cd content in topsoil presented extremely significant positive correlation with soil organic matter and total N, while no significant correlation with pH and total P. Long-term and excessive usage of nitrogen and organic fertilizer is the main reason for Cd accumulation in greenhouse soils. With the increase of planting years, a significant increasing trend is found in soil organic matter and total N, and a little decline trend in soil pH. Cd content in soil increased significantly with the increase of cultivating years due to the universal high input of organic/chemical fertilizers in the study area. In the soil of Cd contamination, the widespread environmental problems such as secondary salinization and acidification may further increase of the proportion of available Cd in greenhouse soil and accordingly enhance the risk of Cd intake by people via eating vegetables.

ACKNOWLEGEMENTS

This research is supported by the National Natural Science Foundation of China (No. 41101472) and Natural Sciences Foundation of Shandong Province (No. 2009ZRB019E5)

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