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Temporal-spatial distribution and variability of cadmium contamination in soils in Shenyang Zhangshi irrigation area, China

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Abstract: Heavy metal contamination in soils has been of wide concern in China in the last several decades. The heavy metal contamination was caused by sewage irrigation, mining and inappropriate utilization of various agrochemicals and pesticides and so on. The Shenyang Zhangshi irrigation area (SZIA) in China is a representative area of heavy metal contamination of soils resulting from sewage irrigation for about 30 years duration. This study investigated the spatial distribution and temporal variation of soil cadmium contamination in the SZIA. The soil samples were collected from the SZIA in 1990 and 2004; Cd of soils was analyzed and then the spatial distribution and temporal variation of Cd in soils was modelled using kriging methods. The kriging map showed that long-term sewage irrigation had caused serious Cd contamination in topsoil and subsoil. In 2004, the Cd mean concentrations were 1.698 and 0.741 mg/kg, and the maxima 10.150 and 7.567 mg/kg in topsoils (0–20 cm) and subsoils (20–40 cm) respectively. These values are markedly more than the Cd levels in the second grade soil standard in China. In 1990, the Cd means were 1.023 and 0.331 mg/kg, and the maxima 9.400 and 3.156 mg/kg, in topsoils and subsoils respectively. The soil area in 1990 with Cd more than 1.5 mg/kg was 2701 and 206.4 hm² in topsoils and subsoils respectively; and in 2004, it was 7592 and 1583 hm², respectively. Compared with that in 1990, the mean and maximum concentration of Cd, as well as the soil area with Cd more than 1.5 mg/kg had all increased in 2004, both in topsoils and subsoils.

Keywords: temporal-spatial distribution; cadmium contamination of soil; Zhangshi sewage irrigation area (ZSIA); Shenyang

Introduction

Sewage irrigation in farmland as a technology for wastewater disposal and agricultural usage was widely applied in many districts in the world (Sun *et al.*, 2001). Now the heavy metal contamination of soils derived from sewage irrigation and the consequent contamination problems of ecological systems and plants derived from contaminated soils have become a focus of worldwide attention. The Shenyang Zhangshi irrigation area (SZIA), China, is a representative area of heavy metal contamination of soils as a result of sewage irrigation.

The irrigation area occupied 28000 hm² and has been used for sewage irrigation for 30 years since 1954 (Chen *et al.*, 1980). Heavy metals had accumulated in soils in irrigation areas, in which the Cd concentration rose from 2.4 to 24.0 mg/kg in soils, and in rice grains rose from 1.0 to 2.0 mg/kg (Chen *et al.*, 1985). Many studies on soil heavy metal contamination in SZIA were carried out after 1970, because of the high Cd concentration detected in rice grain. The land was partly rezoned from crop planting to industry use in 1992. However, currently there are still many areas used for planting vegetables and crops.

Some studies focused on micro-interface processes, contamination mechanism of Cd and appraisal of the ecological effects of Cd contamination in soils (Wu *et al.*, 1986, 1992; Yu, 1991). These studies were based on individual samples only, and did not analyze the temporal-spatial variability and

ecological effects of regional heavy metal contaminations on a large scale.

When evaluating the environmental impacts of heavy metals, the initial determination of its spatial distribution is an important aspect. The spatial distribution and temporal variation of Cd contamination in soils can be determined by geostatistical methods based on sampling at different times (Atteia *et al.*, 1994; Barabás *et al.*, 2001; Bierkens, 1997; Carlon *et al.*, 2001; Goovaerts *et al.*, 1997; Von Steiger *et al.*, 1996). These techniques provide methods to estimate either the value of a soil attribute at locations between samples, or the probability that the attribute value will exceed a given threshold at a particular location. Such information is essential for mapping potential risks to the environment or to human health. Based on the archival information and data in 1990 and the corresponding investigation survey of Cd contaminations in 2004, this paper studied the spatial distributions of Cd in soils and the variation over time in SZIA, China. The resulting conclusions are valuable for the management and remediation of the heavy metal contaminated soils in SZIA and may be applicable to other contaminated areas.

1 Materials and methods

1.1 Study area

The SZIA, located in the western suburb of Shenyang City (Fig.1), is one of the most important heavy industry cities in China and is the political and

cultural centre of Liaoning Province. Shenyang City is located on the alluvial plain of the central part of the Liao River in central Liaoning Province. The study area has an even landform with a slope of 1:1000 and the geographical coordinates are $123^{\circ}11'18''$ — $123^{\circ}20'37''$ E and $41^{\circ}43'14''$ — $41^{\circ}48'41''$ N. The main soil types in SZIA are meadow brown soil, paddy with organic matter of 1% to 2.6%. The pH is from 5.8 to 6.5 and the clay of soils (less than 0.01 mm) is from 24% to 62%. The clay mineral of the soil is mainly illite with a little montmorillonite. The main plants in SZIA are rice, some corns and vegetables. The sewage irrigation area covered 200 hm² in 1954, increased to 28000 hm² in 1968 and irrigation continued for about another 25 years. The irrigation water was the mixture of the surface water of the Hun River (1.0—2.0 m³/s) and the sewage of Weigong Open Ditch (3.5—4.0 m³/s). The sewage contained not only a great deal of organic matters, but also a great deal of heavy metals. The Hun River was almost uncontaminated. The

sewage irrigation in SZIA was ceased in 1992. The land was partly rezoned from crop planting to industry use due to heavy metal accumulation in soil (and also high Cd retained in rice grain) found in 1970.

1.2 Field sampling

Soil samples for different crop cultivations of rice, corn, and vegetable etc. were collected from the topsoil (0—20 cm) and subsoil (20—40 cm) in October 1990 with a regular grid design of 100 hm² spacing in an area of 15400 hm² (Fig.1a). Each soil sample consisted of 4 subsamples weighing 0.2 kg, which were distributed evenly on two diagonal lines of the 100 hm² grid. In October 2004, soil samples for different crop cultivations of rice, corn, and vegetable etc. were collected respectively from the topsoil (0—20 cm) and subsoil (20—40 cm) with a stratified random sampling design in the same area as that in 1990 (Fig.1b), because soils were hardly collected with a regular grid design like that in 1990. The four subsamples were mixed and placed at the center of grid.

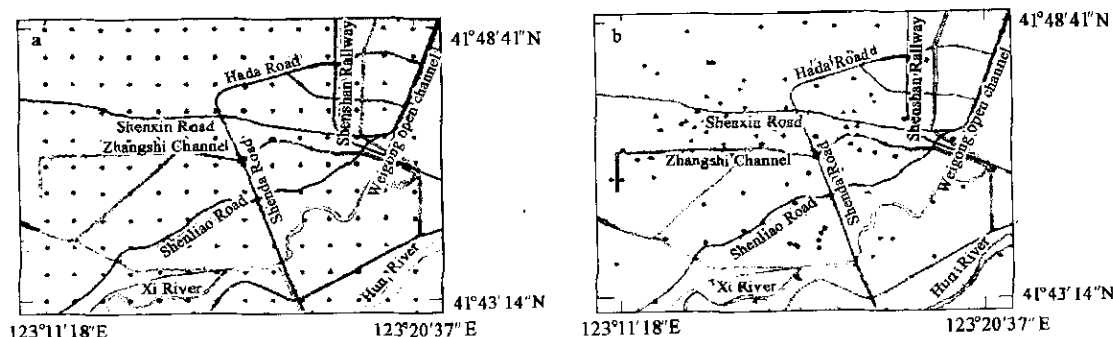


Fig.1 Location map of the study area and sampling points in 1990(a) and in 2004(b)

There were total 154 and 94 samples in 1990 and 2004 respectively. Eleven profile sections (60 cm or 100 cm) were selected and soil samples in these profile sections were collected every 20 cm in 2004. All soil samples were collected in polyethylene bags. The samples were air dried at room temperature and ground with an agate mortar and pestle to pass through a 2-mm stainless steel sieve, then stored at 4°C before analyses.

The soil samples were digested in a heating block with aqua regia in polyethylene pots and Cd analyses were conducted with flame absorption spectrophotometry on a Varian (Melbourne, Australia) Spectr AA-220 with background correction. Analysis quality was monitored with criteria soil of the first grade (GBW07401 and GBW07404) in China according to standard methods of China (GB/T17140-1997 in Liu FZ, 2001). The analysis precision of Cd was 0.002 mg/kg for flame atomic absorption spectrophotometry on a Varian (Melbourne, Australia) SpectrAA-220.

1.3 Weak acid extraction test

1.3.1 Weak acid extraction

According to the extraction method of Loring and Rantala (1992), two grams of dried sediment sample were weighed in a polythene centrifuge tube to which 25 ml of 25% (v/v) HOAc was added. Then, the tubes were shaken mechanically for 6 h, centrifuged for 10 min at 2500 r/min and the clear supernatant was poured into an acid cleaned 50 ml volumetric flask. Then 10 ml of ultra-pure deionized water was added to the solids in the tube, shaken briefly, centrifuged and the supernatant was added to the flask and made up to volume (50 ml) with ultra-pure deionized water. Analysis of nondetrital fraction of Cd was performed by graphite atomic absorption spectrophotometry on a Varian (Melbourne, Australia) SpectrAA-220 too. Analytical blanks and sediment samples were analyzed in triplicate using the same procedures and reagents. The coefficient of variation was 4.5%.

1.3.2 Eluviation test

The test soils were collected from ZSIA. The geographical coordinate of sampling location was $41^{\circ}46'04''$ N and $123^{\circ}16'31''$ E. The soil was sampled, natural air-dried, shattered, and then put in column according to natural stratus. The test installation was

polyethylene column with 5 cm of diameter and 80 cm of length. The column had been marinated for 24 h with 10% HNO₃ solution, then rinsed with deion water, air-dried. The bottom of column was fixupped with filtration membrane, and the filtrate was collected with beaker. The 1500 ml rainwater (pH 7.1) and 1500 ml deion water (pH 6.12) were used to simulate Cd variation of soil profile during rainfall and irrigation based on mean precipitation (700—800 mm/a). The test was performed by three stages in 54 h, and the solution used was 500 ml in every stage. The eluviated soil column was extrated into 6 parts according to 10 cm high stratum on the day 6 after the test was over, then air-dried, ground, digested, and Cd was analyzed. The soil digestion process and Cd analysis method were as above.

1.4 Data processing

The survey of the content and spatial distribution of heavy metals in contaminated soil is very important for assessing their impact on environment and agricultural activities. Geostatistics (Sufer Version 6.04, June 24, 1996, Golden Software, Inc.) provides tools for describing spatial variation of soil properties and for local interpolation (kriging methods) to predict and map values at non-sampled locations. In recent years, geostatistics has been applied in studies on the distribution and environmental behavior of contaminants in soils because it is able to treat the variables of interest as regionalized variables explicitly and

demonstrate their actual spatial distribution pattern (Cattle *et al.*, 2002; Wang and Liu, 2004; Amini *et al.*, 2005). Compared with many other geostatistics methods such as the trend surface analysis, kriging method provides optimal estimates by interpolation (theoretically without bias and with minimal variance), for tracing out pollution of unrecorded sites, based on the theory of regionalized variables (Zhang *et al.*, 2002; Liu *et al.*, 2002; Zhang and Fan, 2002). Kriging provides a more accurate description of the data spatial structure and produces valuable information about estimation error distributions (Avendaño *et al.*, 2003; Litaor *et al.*, 2003). Therefore we used kriging analysis to estimate the spatial distribution pattern of heavy metals in SZIA. The levels of the interpolation curves were determined at (1) 0.110 mg/kg (background levels of soil Cd in the studied district), (2) 0.600 mg/kg (the levels in second grade standard soil in China according to GB15618-1995) and (3) 1.500 mg/kg (2.5 times the Cd level in the second grade standard soil).

2 Results

2.1 Statistical features of soil Cd

The statistical distributions of soil Cd were examined and statistical parameters were calculated with geostatistics methods (Table 1). Statistical distributions were plotted (Fig.2).

The results indicated that the frequency

Table 1 Statistical parameters of soil Cd in SZIA							
Stratum	Year	Mean, mg/kg	Range, mg/kg	Coefficient of variation	Variance	Coefficient of skewness	Coefficient of kurtosis
0—20 cm	1990	1.023	0.179—9.400	1.916	1.217	0.704	14.13
	2004	1.698	ND—10.150	1.637	1.547	0.389	0.608
20—40 cm	1990	0.331	0.100—3.156	5.915	1.217	1.649	0.135
	2004	0.741	ND—7.567	4.735	1.983	0.278	0.593

Note: ND. no detected

distribution and the logarithmic frequency distribution of soils Cd concentration were clearly nonnormal. In 2004 for topsoils and subsoils, the coefficients of kurtosis were 0.608 and 0.593 respectively, and the coefficients of skewness were 0.389 and 0.278 respectively. For 1990's data the coefficients of kurtosis were 14.13 and 0.135, the coefficients of skewness were 0.704 and 1.649 respectively. These large deviations from Gaussian distributions indicated anthropogenic influence on soil Cd. In 2004, the Cd means were 1.698 mg/kg (0—20 cm) and 0.741 mg/kg (20—40 cm), which were respectively 2.83 times and 1.24 times the level in second grade standard soil (LSGSS) in China. The ranges between minimum and maximum (ND to 10.150 mg/kg in topsoils and ND to 7.567 mg/kg in subsoils), the coefficient of variation (1.637 in topsoils and 4.735 in subsoils) and variance (1.547 in topsoils and 1.983 in subsoils) were

relatively high, revealing heterogeneity of the soil Cd. In 1990, the Cd mean was 1.023 mg/kg (0—20 cm) and 0.331 mg/kg (20—40 cm), which were 1.71 times and 0.55 times the LSGSS in China. The range between minimum and maximum was from 0.179 to 9.400 mg/kg (in topsoils), and from 0.100 to 3.156 mg/kg (in subsoils), the coefficient of variation was 1.916 (in topsoils) and 5.915 (in subsoils), and variance all was 1.217 (in topsoils and in subsoils). The relatively high values of concentration range and coefficient of variation indicated heterogenesis of soil Cd too, although the Cd mean in subsoil was less than the LSGSS in China. 2.2 Spatial distributions of cadmium in soils The data of soil Cd was processed by kriging interpolation in the study area for 1990 and 2004, and results presented the relative areas of contamination for uncontaminated soils (Cd < 0.110 mg/kg), second

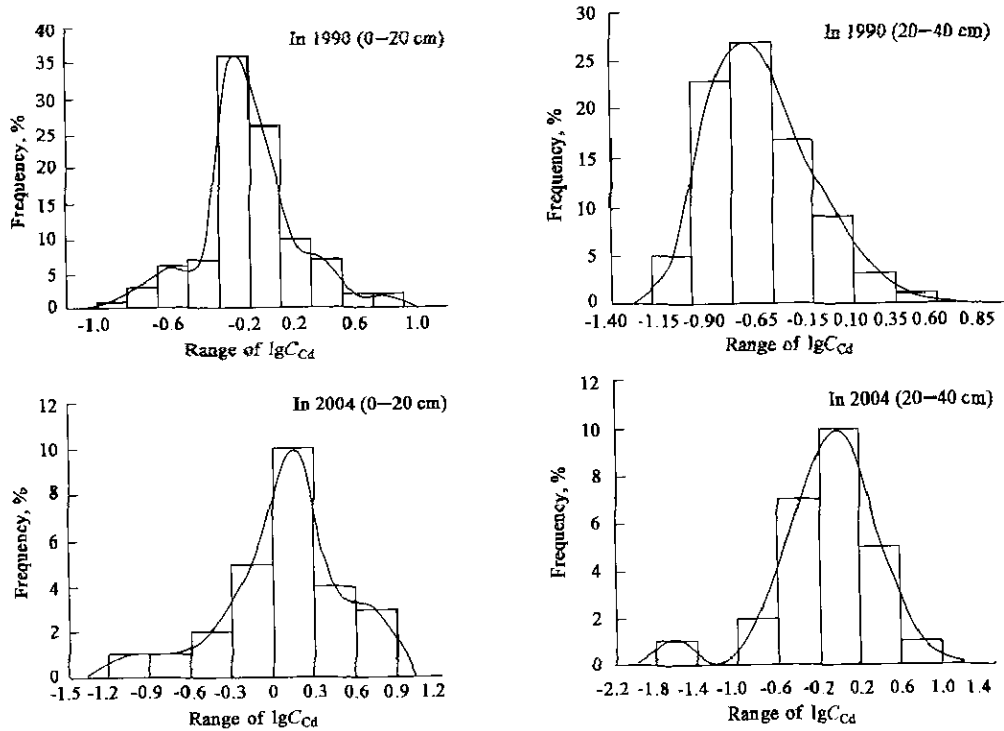


Fig.2 Logarithmic frequency distribution of soil Cd

grade soil (Cd, 0.110–0.600 mg/kg) and contaminated soils (Table 2). The contour maps of Fig.3 and Fig.4 show the spatial distributions of Cd contamination in topsoil and the subsoil for 1990 and 2004.

In 1990 (Fig.3a), the topsoil area with Cd ranged from 0.110 to 0.600 mg/kg was 2821 hm², which was

18.3% of soil area studied. The soil area with Cd from 0.600 to 1.500 mg/kg was 9878 hm² (64.1%). The soil area with Cd more than 1.500 mg/kg was 2701 hm² (17.5%) and there was no soil with Cd less than 0.110 mg/kg (that is the background level of Cd in studied area).

In 2004 (Fig.3b), the topsoil area with Cd

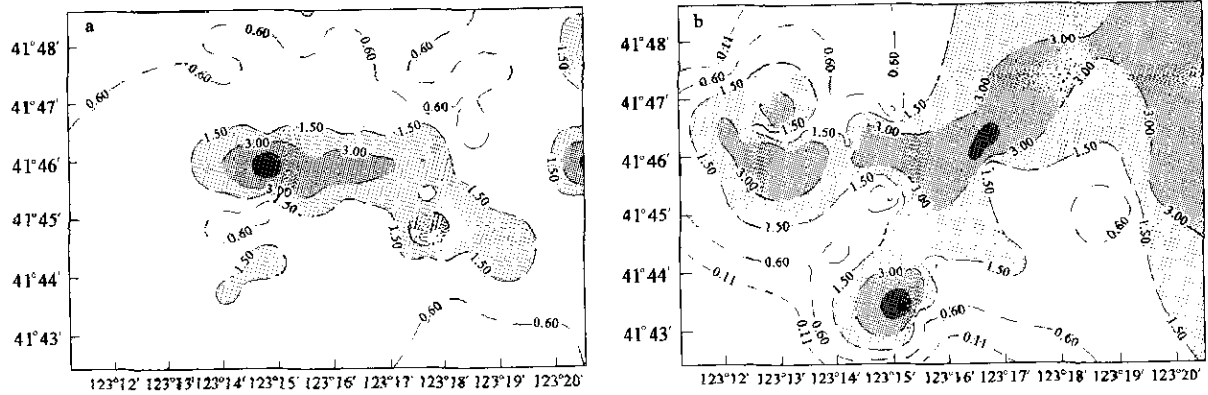


Fig.3 Spatial distributions of Cd in topsoils in 1990 (a) and in 2004(b)

between 0.110 and 0.600 mg/kg was 1996 hm² (13.0% of the study area). The soil area with Cd between 0.600 and 1.500 mg/kg was 4594 hm² (29.8%). The soil area with Cd more than 1.500 mg/kg was 7592 hm² (49.3%) and the area with Cd less than 0.110 mg/kg was 1218 hm², which was 7.9% of the study area.

In 1990 (Fig.4a), the subsoil area with Cd between 0.110 and 0.600 mg/kg was 6180 hm², which was 40.3% of the study area. The soil area with Cd between 0.600 to 1.500 mg/kg was 1066 hm² (6.9%).

The soil area with Cd more than 1.500 mg/kg was 206.4 hm² (1.4%) and the area with Cd less than 0.110 mg/kg was 7920 hm² (51.4%).

In 2004 (Fig.4b), the subsoil area with Cd between 0.110 and 0.600 mg/kg was 6389 hm², which was 41.5% of the study area. The subsoil area with Cd between 0.600 to 1.500 mg/kg occupied 4806 hm² (31.2% of the study area). The subsoil area with Cd more than 1.500 mg/kg occupied 1583 hm² (10.3% of the study area) and the subsoil area with Cd less than 0.110 mg/kg was 2621 hm² (17.0% of the study area).

Table 2 Extents of soils at different Cd levels in study area (as a percentage of the total sampled area)

Cd level, mg/kg	In topsoil, %		In subsoil, %	
	1990	2004	1990	2004
<0.110	0.00	7.90	51.4	17.0
0.110—0.600	18.3	13.0	40.3	41.5
0.600—1.500	64.2	29.8	6.90	31.2
>1.500	17.5	49.3	1.40	10.3

2.3 Distribution of soil cadmium in vertical profile sections

Fig.5 gives the distribution curves of Cd in different vertical soil profile sections in 2004. The curves show that, for a background vertical profile section (Section 1), the Cd level was nearly constant from top to bottom, and yet for polluted vertical profile sections (Sections 2—4), the Cd level was markedly augmented in topsoils and evidently decreased with depth of soil, indicating anthropogenic Cd in topsoils.

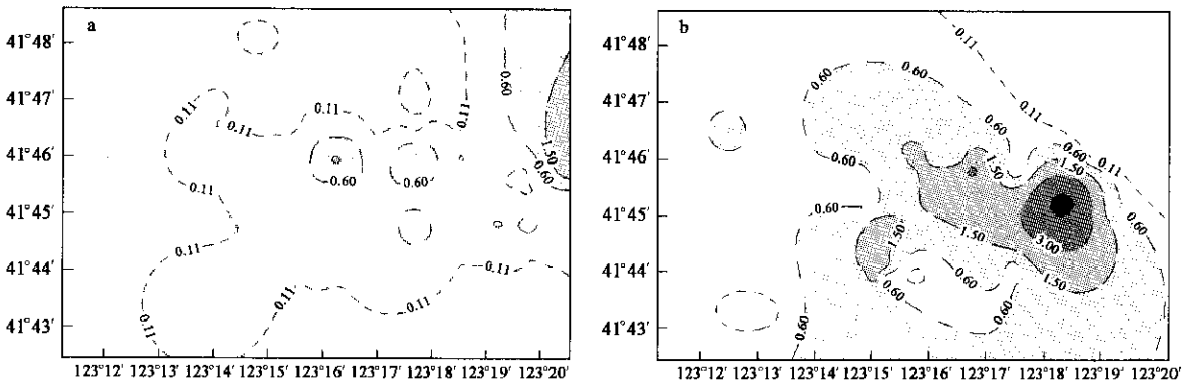


Fig.4 Spatial distributions of Cd in subsoils in 1990 (a) and in 2004 (b)

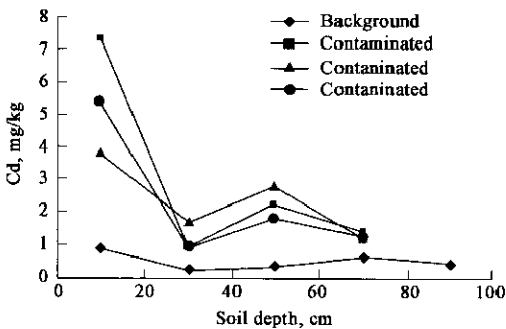


Fig.5 Cd distribution in soil vertical profile in sections

2.4 Temporal-spatial variations of cadmium in soils

The mean and the maximum concentration of Cd increased from 1.023 and 9.400 mg/kg in 1990 to 1.698 and 10.150 mg/kg in 2004 for topsoils, and from 0.331 and 3.156 mg/kg in 1990 to 0.741 and 7.567 mg/kg in 2004 for subsoils (Table 1). The soil extent with Cd more than 1.500 mg/kg also increased, from 2701 hm² in 1990 to 7592 hm² in 2004 for topsoils, and from 206.4 hm² in 1990 to 1583 hm² in 2004 for subsoils(Figs.3 and 4).

3 Discussion

3.1 Data usability

Although the location and number of soil sampling were different between in 1990 and in 2004, soil samples were collected with the same sampling methods in the same soil stratum. In addition, soil

samples were quantitatively adequate for representative of soils because of homogeneous soil type and even landform in the study area. Therefore, the geometry mean and the maximum of Cd concentration were comparable. The data of soil Cd was processed by kriging interpolation in the study area for 1990 and 2004. So the spatial distributions of Cd for 2004 and 1990 were comparable.

3.2 Genetic discussion

The increase of Cd concentration and soil extent contaminated by Cd indicated the Cd import since the sewage irrigation ceased in SZIA in 1992, which may be related to Cd reactivation of the sediments in Xi River and the transportation of contaminated soil by people due to industry development in SZIA. The Xi River, the inland river of Shenyang City, comes from the south of Weigong Open Ditch, and carries the sewage and industrial waste water at all times. The water monitoring from different section of Xi River presented that Cd contents in water were 0.010 to 0.069 mg/kg in 1990, which was 2 to 13.8 times of the criteria of agricultural irrigation water in China(0.005 mg/kg), and the Cd contents were 0.005 to 0.040 mg/kg in 2004, which was 1 to 8 times of the criteria. The sediments of Xi River were also analyzed, and the results indicated that Cd contents were 1.13 to 15.73 mg/kg in 2004. The available Cd (acetic acid-HOAc, 25%(v/v)) of sediments is weakly held in ion exchange sites, held easily in soluble amorphous compounds of iron and manganese, carbonates and those

metals weakly held in organic matter (Selvaraj *et al.*, 2004) and is operationally defined as nondetrital (acid soluble). The available Cd is from 37.4% to 52.0% of total Cd. These features showed that Cd in sediments may be reactivated into water, then be carried into soils by irrigation water.

The transportation of contaminated soil by people due to industry development in SZIA also partly explained the variation of higher Cd extent in topsoils.

The soil extent with Cd ranged from 0.600 to 1.500 mg/kg decreased in topsoils, while increased in subsoils over the same time period. Those features may be related to the eluviation of Cd in contaminated topsoils with rainfall and irrigation water and redeposition in subsoils because of the comparative acidity of topsoils (pH 5.79) compared with subsoils (pH 6.12). The eluviation test of soil column (pH: 7.15, 6.12; soil column height: 60 cm; leach time: 72 h) for soils contaminated with Cd were carried out in order to investigate the variation of soil Cd in profile. The results indicated that 24% Cd was leached in the topsoil (0–30 cm) and was redeposit in subsoil (30–60 cm) in pH 7.15, and that 40% Cd in topsoil and 62% Cd in subsoil were leached in pH 6.12, which explained Cd variation in topsoils and subsoils.

4 Conclusions

Sewage irrigation over a long time had caused serious Cd contamination in soil in SZIA, China. At present, the mean of soil Cd is markedly higher than the LSGSS in China, and the maximum of soil Cd found in this study was 16.92 times of the LSGSS in China. The mean of soil Cd and the soil area with high Cd concentration (more than 1.500 mg/kg) had evidently increased since sewage irrigation ceased in 1992, both in topsoils and in subsoils. The soil extent of Cd contamination augmented in topsoils and subsoil too. Those features were mainly related to Cd reactivation of contaminated sediments in Xi River and Cd importment during irrigation and the eluviation of Cd in contaminated topsoils with rainfall and irrigation water.

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