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Assessment of Heavy Metal Pollution and Potential Ecological Risk in Soils of Tianjin Sewage Irrigation Region, North China

Beibei Hu, Jun Zhou, Lidong Liu, Weiqing Meng and Zuwei Wang*

College of Urban and Environment Science, Tianjin Normal University, Tianjin 300387, People's Republic of China

Abstract

The aim of the study was to investigate heavy metals concentrations in wheat field soils of Tianjin sewage irrigation region, and evaluate the potential ecological risk of heavy metals pollution. ICP-OES was employed to analyze the heavy metals concentrations in wheat field soils of Tianjin sewage-irrigation region. The grading standard of Tianjin soil environmental quality was used as the assessment foundation for soils and Lars Hakanson's potential ecological hazards index method was used to evaluate the heavy metals pollution potential ecological risk in soils. The results showed that: Cd concentrations in soils ranged from 0.03 mg•kg^{-1} to 1.17 mg•kg^{-1}, with an average value of 0.46 mg•kg^{-1}; Zn concentrations in the soils ranged from 62 mg•kg^{-1} to 307 mg•kg^{-1}, with an average value of 129.08 mg•kg^{-1}. The concentrations of Cd and Zn in the soils exceeded second grade standards of Tianjin soil environment quality, so the soil was polluted. The other heavy metal elements, such as Cu, Pb, Cr and Ni, were all lower than second level of Tianjin soil environment quality standards, meaning that the soil was not contaminated by these heavy metals. The heavy metal concentrations were relatively rich in saline wet fluvo-aquic soil and clayification fluvo-aquic soil. Based on Lars Hakanson's potential ecological hazards index method, Cd had heavily ecological risk in soils and was the main pollutant, while the other heavy metals had lightly ecological risk. Ecological risk of total heavy metals pollution was moderate. The heavy metals were most likely from wastewater irrigation. The pollutants in the soils came mainly from sewage irrigation. The waste water treatment technology should involve steps to remove heavy metals causing risk to human health.

Keywords: Heavy metal pollution; Potential ecological risk; Pollution source; Soil; Tianjin sewage irrigation region

Introduction

Heavy metal is one of the important pollutants in the environment as a result of both natural and anthropogenic activities. Wastewater irrigation, solid waste disposal, sludge applications, vehicular exhaust and industrial activities are the major sources of soil contamination with heavy metals, and an increased heavy metal uptake by food crops grown on such contaminated soils is often observed. Wastewater irrigation is a widespread practice in the world and recently a number of articles have been published on wastewater-irrigated soils contaminated with heavy metals [1-11].

Heavy metals have a potential to contaminate soil, which can be dispersed and accumulated in plants and animals, and taken in by humans through consumption [12,13]. So heavy metals contamination has been a worldwide environmental concern with its potential ecological effect [14,15]. The present work deals with the quantification of heavy metal concentrations in soil and crops grown in Tianjin sewage irrigation region, the biggest sewage irrigation region in China which has 25-40 years sewage irrigation history. In earlier studies heavy metal concentrations were discussed [16-19] and the impact of heavy metals pollution on vegetables and crops was assessed [20-25]. While literature on the heavy metal pollution in wheat field soil of Tianjin sewage irrigation region was rare. Wheat is one of three main foods in north China and is main crop in Tianjin sewage irrigation region, so it is important to assess the potential ecological risk of heavy metals in wheat field soils in the study area for human health.

Soils of Tianjin Sewage Irrigation Region were examined with the following objectives: (1) to measure the concentrations of selected metals (i.e., Cd, Cu, Pb, Zn, Cr and Ni) in soil; (2) to assess the potential ecological risk of heavy metals in soil; and (3) to identify pollution source of heavy metals in soil. The results of this research are needed for the government to control heavy metal pollution.

Materials and Methods

Study area

Tianjin wastewater irrigation region includes three main sewage discharge channel systems; they are North sewage discharge channel region, South sewage discharge channel region and Beijing sewage discharge channel region. The history of sewage irrigation was 25-40 years.

The North sewage discharge channel region is located in Dongli District with an irrigation region of 1.2 × 104 hm^2. The South sewage discharge channel region is located in Xiqing District and Jinnan District with an irrigation region of 2.33 × 104 hm^2. Beijing sewage discharge channel region in Tianjin is mainly located in Wuqing District and Beichen District, with an irrigation region of 3.87 × 104 hm^2 in Wuqing District and 0.95 × 104 hm^2 in Beichen District. Over the past four decades, human activities and prolonged use of irrigation water mixed with sewage have resulted in soil pollution with heavy metals of the study area [26].

The study area is located in the temperate zone continental climate, and the annual average temperature for the area is 11.4 - 12.9°C average

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***Corresponding author:** Zuwei Wang, College of Urban and Environment Science, Tianjin Normal University, Tianjin 300387, People's Republic of China, Tel: +8613512014515; E-mail: hubeibei0328@163.com

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annual precipitation 520 - 660 mm and the annual relative humidity in the range of 60% - 80%. Tianjin sewage irrigation region is plain terrain, where the soil types are mainly fluvaquents [27], which named as Ustochrepts by USA and Cambisols by UN. There are four types of fluvo-aquic soils in the study area, including ordinary fluvo-aquic soil, saline fluvo-aquic soil, wet fluvo-aquic soil and saline wet fluvo-aquic soil. Ordinary fluvo-aquic soil was main soil type. The main crops include winter wheat, summer corn and vegetables.

Soil sampling and analysis

Soil sampling: For this study, a total of 98 soil sampling sites were selected by using grid method (Figure 1). Using the quincunx distribution point method, each topsoil sample was collected from the plow layer (0 - 20 cm) at the central point and the four apexes of a 5 km × 5 km square. Approximately 1 kg of soil was collected, respectively, and then mixed, using the quartering method. All soil samples were air-dried, sieved to <2 mm, homogenized, and stored in Kraft paper envelopes prior to laboratory analysis and processing.

Soil samples analysis and quality control: Soil samples were analyzed for pH values, organic matter contents, total soil concentrations and the concentrations of main heavy metal pollutants, such as Cd, Cu, Pb, Zn, Cr and Ni.

The soil pH values were measured by the voltammetric titrate method (PHs-10A) with a soil/water ratio of 1: 2.5. The organic matter concentrations of soil were measured by the $K_2Cr_2O_7$ method. The soil salinity was measured gravimetrically. For extraction of heavy metals such as Cd, Cu, Pb, Zn and Cr, one gram dried soil was digested in 15 ml mixture of HNO_3, H_2SO_4 and $HClO_4$ (5:1:1) at 80°C until a transparent solution was obtained [28]. Water samples (50 ml) were digested with 10 ml of concentrated HNO_3 at 80°C until the solution became transparent. These transparent solutions were then filtered through Whatman number 42 filter papers and diluted to 50 ml with distilled water. The concentrations of Cd, Cu, Pb, Zn and Cr in the filtrate were determined by using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES, Leeman Labs, USA), fitted with a specific lamp of particular metal using appropriate drift blanks. The experiment was repeated three times.

The blank reagent and standard reference soil (from the National Research Center for Standards in China) were included in each sample batch to verify the accuracy and precision of the digestion procedure and subsequent analyses. Repeated analyses of standard samples were regularly carried out to control reproducibility.

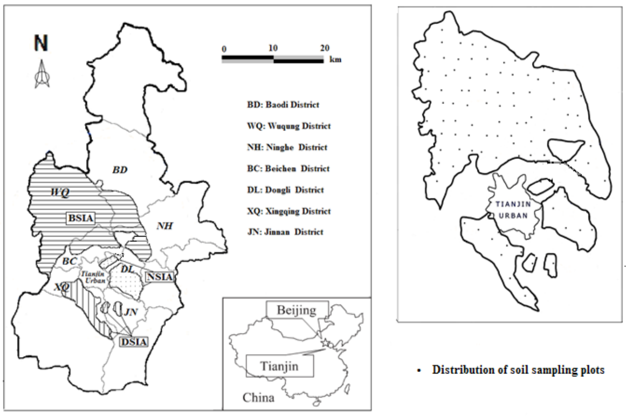

Figure 1: Location of study area and sampling sites.

Contamination assessment methodology

The potential ecological risk index method, which is based on the characteristics of a heavy metal and its environmental behavior, was advanced by the Swedish scholar Lars Hakanson [15,29-31], according to the characteristics of heavy metal and its environmental behaviour. The potential ecological risk index method is an approach to evaluate the heavy metal contamination from the perspective of sedimentology. It not only considers the heavy metal level in the soil, but also associates the ecological and environmental effects with toxicology and evaluates the pollution level using the comparable and equivalent property index grading method. The potential ecological risk index is related to the individual pollution coefficient, the heavy metal toxicity response coefficient, and the potential ecological risk individual coefficient, and is expressed as follows [29,31].

$$R = \sum_{i=1}^{m} E_r^i = \sum_{i=1}^{m} T_r^i \bullet C_f^i = \sum_{i=1}^{m} T_r^i \frac{C^i}{C_n^i}$$

Where E_r^i is the potential ecological risk individual coefficient and T_f^i is the toxicity response coefficient of a certain kind of metal toxicity using the standard heavy metal toxicity coefficient developed by Hakanson as a reference. The values for each element are in the order of Zn=1<Cr=2<Cu=Ni=Pb=5<As=10<Cd=30<Hg=0.

C_f^i is the individual pollution coefficient, C^i is the actual observed value of heavy metals in surface soil, and C_n^i is a relative ratio, which is obtained from the background values of soil heavy metals in Tianjin. The detailed grading standard of heavy metal individual pollution coefficients and pollution ecological risk coefficients are listed in Table 1 [29,31].

Correlation analysis

The correlation analysis was selected to identify pollution sources [32] and the data in this work were performed with SPSS 12.0 for Windows [33].

Results and Discussion

Physicochemical properties and metal levels of soil

The pH value in the upper layer soils ranged from 7.3 to 8.4, with an average value of 7.62, meaning that the soil was alkaline. The organic matter concentrations in the upper layer soils ranged from 0.80% to 2.7%, with an average value of 1.47%. The soil salinity in the upper layer soils ranged from 0.03% to 0.144%, with an average value of 0.064%, and part of the soils belonged to salinization soil.

The descriptive statistic of metal levels in upper horizon soils in Tianjin sewage irrigation region was summarized in Table 2. In addition, the environmental quality standards for soils of Tianjin were used as the reference values [34].

In general, the concentrations of heavy metals varied widely in the studied region. The mean contents of Cd and Zn in soils exceed the reference values. 89.8% of the soil samples exceeded obviously the second Grade standard of Cd in Tianjin soil environmental quality; further analysis showed that 75% of the soil samples exceeded the third Grade standard and 34.9% exceeded the fourth Grade standard of Cd in Tianjin soil environmental quality, so the soil were moderately polluted by Cd. 46.94% of soil samples exceeded the second level of Zn in Tianjin soil environmental quality and 34.78% of soil samples exceeded the third level, so the soil had been lightly contaminated by Zn. The average concentrations of Cd and Zn in soil from other studies [18,19] were 0.24-1.86 mg·kg^{-1} and 129-142 mg·kg^{-1}, which were similar

E_r^i	**Grades of ecological risk of single metal**	R_i	**Grades of potential ecological risk of the environment**
$E_r^i < 40$	light	$R_i < 150$	light
$40 \le E_r^i < 80$	moderate	$150 \le R_i < 300$	moderate
$80 \le E_r^i < 160$	heavy	$300 \le R_i < 600$	heavy
$160 \le E_r^i < 320$	severe	$R_i \ge 600$	severe
$320 \ge E_r^i$	Very severe		

Table 1: Indices and grades of potential ecological risk of toxic metals contamination.

Element	**Cd**	**Cu**	**Pb**	**Zn**	**Cr**	**Ni**
Ranges (mg·kg^{-1})	0.05-1.17	10.92-61.32	3.8-49.79	62.22-333.6	40.16-108	15.91-61.42
Average (mg·kg^{-1})	0.46	28.15	15.62	129.08	64.19	32.08
SD	22.29	9.45	6.34	56.23	12.25	7.42
CV (%)	48	33.58	40.57	43.57	19.09	23.14
Tianjin Standard- Class I a	≤ 0.09	≤ 28.35	≤ 20.32	≤ 76.27	≤ 98.38	≤ 32.39
Tianjin Standard- Class II b	>0.09 ≤ 0.159	>28.35 ≤ 43.71	>20.32 ≤ 32.83	>76.27 ≤ 115.3	>98.38 ≤ 107	>32.39 ≤ 46.35
Tianjin Standard- Class III c	>0.159 ≤ 0.380	>43.71 ≤ 70	>32.83 ≤ 190	>115.3 ≤ 190	>107 ≤ 228	>46.35 ≤ 53
Tianjin Standard- Class IV d	>0.380 ≤ 0.6	>70 ≤ 100	>190 ≤ 350	>190 ≤ 300	>228 ≤ 350	>53 ≤ 60
Tianjin Standard- Class V e	>0.6 ≤ 1.0	>100 ≤ 400	>350 ≤ 500	>300 ≤ 500	>350 ≤ 400	>60 ≤ 200
Tianjin Standard- Class VI f	>1.0	>400	>500	>500	>400	>200

Note: a. Environmental quality standard for soils in Tianjin (Environmental Monitor Center of Tianjin, 1990), and numbers in class I are threshold levels of natural background value in Tianjin; b. Metal levels in class II are meaning the soil is clean. It is used as the soil assessment criterion; c. Metal levels in class III are meaning the soil is lightly polluted by heavy metals; d. Metal levels in class IV are meaning the soil is moderately polluted by heavy metals; e. Metal levels in class V are meaning the soil is heavily polluted by heavy metals; f. Metal levels in class VI are meaning the soil is severely polluted by heavy metals.

Table 2: Descriptive statistics of metal levels in soils.

with this paper. It was implied that Cd and Zn may pose potentially unfavorable effects to crops and human health and the remarkable accumulation of them in soils indicated the significant anthropogenic inputs of pollutants.

The concentrations of Cu, Pb, Cr and Ni in most samples were lower than the second standard, and only six Cu soil samples and three Pb soil samples exceeded the second Grade standard, meaning that the soils were not contaminated by Cu, Pb, Cr and Ni.

The concentrations of heavy metals are affected by physicochemical properties of the soils. Their correlation was shown in Table 3.

Results showed that moderate correlation existed between the concentrations of heavy metals and the organic matters in the soil. So, the higher concentrations of organic matters of soil could lead to the bigger absorbance for heavy metals.

The order of the correlation between heavy metals and soil organic matters was Cu>Cr>Pb>Cd>Ni>Zn. The reason was that organic matters contained a large number of functions such as carboxyl, phenolic-based and alcohol-based groups, which could synthesize with heavy metals ions under certain conditions [28,35,36].

There was a certain degree of positive correlation between heavy metals and soil salinities, and the order of the correlation was Cr>Cu>Pb>Zn>Cd>Ni. Other researcher got the same conclusions [4,5].

In general, there was moderate correlation between pH values and the heavy metal contents of soils [1]. But in the study area, there was not. The reason might be that the soil was partial alkaline, and pH value ranged from 7.2 to 8.3.

Cd and Zn were main pollutants. The concentration of Cd and Zn in different soil types was shown in Table 4. Additionally, the ordinary fluvo-aquic soil subgroup was selected to study the influence of soil texture, with a focus on sandy fluvo-aquic soil, loamy fluvo-aquic soil and clayey fluvo-aquic soil.

Results showed that there was the highest Cd concentration in the saline wet fluvo-aquic soil, followed by wet fluvo-aquic soil, saline fluvo-aquic soil and ordinary fluvo-aquic soil, respectively. It should be noted that the standard deviation of Cd was relatively small for

Soil property	Cd	Cu	Pb	Zn	Cr	Ni
Organic matter %	0.3595**	0.8109**	0.5450**	0.3159**	0.5765**	0.3527**
Salinity %	0.2631*	0.4294**	0.3958**	0.2691**	0.4370**	0.2018
pH value	-0.0930	-0.1916	-0.1653	-0.1282	-0.2457*	0.0882

**Correlation is significant at the 0.01 level; *Correlation is significant at the 0.05 level.

Table 3: Correlation between the concentrations of heavy metals and physicochemical properties of soils.

Soil Types		Fluvo-aquic Soil Subgroup				Fluvo-aquic Soil Texture		
		Ordinary fluvo-aquic soil (n=48)	Saline fluvo-aquic soil (n=12)	Wet fluvo-aquic soil (n=14)	Saline wet fluvo-aquic soil (n=19)	Sandy fluvo-aquic soil (n=13)	Loamy fluvo-aquic soil (n=22)	Clayification fluvo-aquic soil (n=14)
Cd	Range	0.05-0.77	0.12-0.97	0.11-0.87	0.28-1.17	0.05-0.6	0.05-0.65	0.09-0.77
	Average	0.38	0.42	0.54	0.65	0.33	0.36	0.44
Zn	Range	62.22-333.6	68.92-191.6	75.44-307.2	78.14-232.9	72.38-333.6	75.01-255.8	62.22-266.4
	Average	125.41	115.52	157.49	122.82	148.36	113.44	122.89

Table 4: The concentrations of Cd and Zn in different types of soils (mg·kg^{-1}).

		Cd	Cu	Pb	Zn	Cr	R_i
E_r^i	**Ranges**	16.67-323	1.93-10.81	0.94-12.25	4.07-21.87	0.43-1.09	24.04-369.02
	Average	155.58	4.96	3.84	8.46	0.65	173.49

Table 5: Potentially ecological risk assessment results of toxic metals in soils.

these four subgroups, which could indicate the Cd concentrations in the soils were correlation with the soil salinity.

The average concentration of Zn in the soils from highest to lowest was wet fluvo-aquic soil, ordinary fluvo-aquic soil, saline wet fluvo-aquic soil and saline fluvo-aquic soil, which could indicate the Zn concentrations in the soils were low correlation with the soil salinity.

The concentrations of Cd showed an increasing trend with the content of clay minerals in the soil. The concentrations of Zn showed a decreasing trend with the content of clay minerals in the soil.

Potential ecological risk assessment

The ecological risk assessment results of toxic metals in soils were summarized in Table 5. It was found that the risk indices (E_r^i) of metals were ranked in the following order: Cd>Zn>Cu>Pb>Cr.

The average ecological risks of Cd in soils were above 80, indicating that Cd posed a heavy risk to the local ecosystem. The values of E_r^i for the other metals were below 40, indicating light risk.

In order to quantify the overall potential ecological risk of observed metals in soils, the value of R_i was calculated as the sum of the all five risk factors. It ranged from 24.04 to 369.02, with an average of 173.49, indicating the moderate potential ecological risk. The element of Cd accounted most of the total risks, and the average percentage was 89.68%.

The result could be confirmed by other studies. The heavy metals pollution risk of vegetable field in Tianjin suburbs was evaluated based on Monto-Carlo model by Bin et al. and Shi et al. [18,19], and the result showed that Cd accumulated in soil and vegetable. The source and enrichment of heavy metals in sewage-irrigated region soil of Dagu sewage discharge channel were discussed by Ref. [17]; it was found that the main pollutant in farmland field soil was Cd.

Pollution source of heavy metals

The correlation among heavy metals can be used to speculate their source. For investigation of inter-metal relationships in soils, correlation analysis was conducted, and the correlation matrix was shown in Table 6.

Results showed that positive correlation existed among Cd, Cu, Pb, Zn, Cr and Ni except the correlation between Zn and Ni. Zn could enter into soils by fertilizer use [37]; so the correlation between Zn and other heavy metals was lower. The highly positive correlation among soil heavy metals suggested that these heavy metals had similar pollution sources. Therefore, the positive correlation among heavy metals in soils of the study area might indicate the combined soil pollution by multi-heavy metals as a result of the rapid development of industry and agriculture as well as human activities, especially sewage irrigation.

Source of heavy metals pollutants in soils could be determined by the study of heavy metals concentrations in soils by different irrigation types. There were four types of irrigation and the order of wastewater irrigation intensity was Wastewater irrigation>Clean water and wastewater mixed irrigation>Intermittent wastewater irrigation>Clean water irrigation. The order of heavy metals concentrations in soils by different irrigation types was similar (Figure 2). It might indicate that pollutants in the soils came mainly from sewage irrigation.

The concentrations of Cd and Zn in waste water was 0.6 mg·L^{-1} and 0.19 mg·L^{-1} separately [38], which was 2 times and 31 times as in the clean water, so they could reach higher concentrations after years of accumulation in soils.

Conclusions

Cd and Zn were the main pollutants of upper horizon soils in the Tianjin sewage irrigation region. However, the soils were not contaminated by other heavy metals, such as Cu, Pb, Cr and Ni. Other researchers acquired the same results [17-19].

The concentrations of heavy metals in soils were influenced by the physicochemical properties of the soils. The organic matter

	Cd	Cu	Pb	Zn	Cr	Ni
Cd	1					
Cu	0.5699**	1				
Pb	0.4703**	0.5873**	1			
Zn	0.3160**	0.3121**	0.2824**	1		
Cr	0.6029**	0.7811**	0.4728**	0.2841**	1	
Ni	0.5775**	0.5545**	0.3022**	0.1840	0.6399**	1

**Correlation is significant at the 0.01 level

Table 6: Pearson correlation coefficients of metal contents in soils.

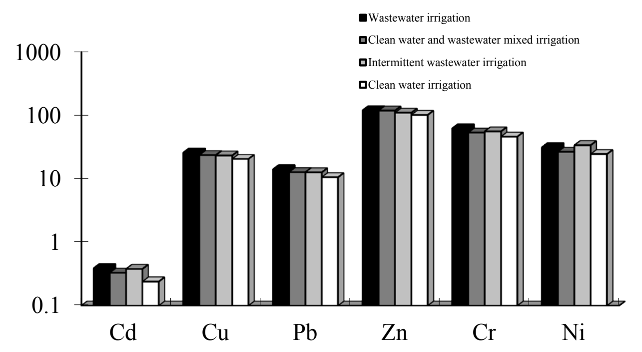

Figure 2: Heavy metals concentrations in soils by different wastewater irrigation intensity (mg·kg^{-1}).

concentrations and salinity of the soils might influence the heavy metal contents of soils [4,5,28,35,36,39]. The pH values of the soils were not correlated with heavy metals concentrations, most likely due to the partial alkalinity of the soils and the narrow pH ranges.

Types and textures of soils had an effect on the heavy metal concentrations and heavy metals were relatively enriched in saline wet fluvo-aquic soils and clayey fluvo-aquic soils. The ecological risk assessment results showed that Cd was the only metal posing a potentially heavy risk to environment while other heavy metals had lightly ecological risk. The overall risk indexes caused by the five toxic metals in soils samples were 173.9, corresponding to moderate risk.

By means of comparison of metal concentrations and correlation analysis, the heavy metals came mainly from wastewater irrigation. Thereby, the control of pollution in wastewater is vital for the mitigation of toxic metal contamination in study area. Additionally, a well-managed waste disposal system is necessary to controlling over the moderate pollution caused by potentially toxic metals.

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