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Experimental Study on the Heavy Metal Pollution in the Soil Irrigated by Reclaimed Water from Sewage Treatment Plant

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Abstract Reclaimed water irrigation is one of the potential ways of solving the shortage of water resources, and the pollution risk on migration behavior of heavy metals in the soil which are irrigated by reclaimed water and the related soil surface is still short of research. Through the experimental study of different kinds of water irrigation methods on vegetable, it can be concluded that compared with sewage irrigation and tap water irrigation, reclaimed water irrigation does not pollute the soil, and it greatly saves the cost of water resources, and even provides a large number of growth elements for vegetables. The results show that after leaching by reclaimed water for 60 days, Cr, Cd, As, Hg, Pb, Ni, Zn and Cu from reclaimed water are enriched in soil to a certain degree, but with the leaching time extending, concentrations of the heavy metal remain stable. The variation of heavy metal content in soil irrigated by reclaimed water is small in vertical depth, basically showing a horizontal trend. According to *Soil Environmental Quality Standards* (GB15618–1995), soil irrigated by reclaimed water does not exceed the standard, better than soil quality standard of planting vegetables.

Key words Reclaimed water, Soil, Vegetables, Accumulation

1 Introduction

There are dense rivers, abundant water resources and fertile lands in Guangzhou City which becomes an important base for the production of high quality and efficient vegetables^[1]. However, with the industrial and agricultural development and population growth, the water pollution is becoming more and more serious^[2–4]. The reclaimed water is the non-potable water reaching certain water quality standards and reused within a certain range after the municipal sewage treatment, so using reclaimed water for irrigation will be a potential way to solve shortage of water^[5–7]. Zhou Lubo *et al.*^[8] study the influence of reclaimed water irrigation in golf course on the groundwater, soil, landscape quality, maintenance cost and human health, and point out that the reclaimed water irrigation in golf course is safe and feasible. There are many foreign studies on farming irrigation by sewage. Lahaam *et al.*^[9] research the impact of different mixing ratio of clean water and reclaimed water on tomato quality; Al-Nakshabandi *et al.*^[10] use reclaimed water for drip irrigation on eggplant near Amman, Jordan; Pollice *et al.*^[11] use the reclaimed water through tertiary sewage treatment for drip irrigation on fennel and tomatoes. The studies show that the reclaimed water as irrigation water is basically safe^[12–13]. However, there is still a shortage of studies on the migration of heavy metals in the soil, the risk of contamination of shallow groundwater and heavy metal accumulation in soils in the process of using reclaimed water to irrigate vegetables^[14–15]. On the basis of long-term wastewater treatment and application by Guangzhou

Sewage Purification Co., Ltd., and Vegetable Research Institute of Guangdong Academy of Agriculture Sciences, we assess the vertical migration of heavy metals in the soil and pollution-related risks in the course of using reclaimed water from Guangzhou's municipal sewage treatment plant to irrigate vegetables, in order to provide the most effective irrigation water for the planting of vegetables in Guangzhou City.

2 Materials and methods

2.1 Materials

2.1.1 Greenhouses. Greenhouse uses galvanized steel frame, with length × width × height of 30000 mm × 6000 mm × 3000 mm. A 2000 mm × 2500 mm door is set at the side of greenhouse. Steel frame is fastened with rivets, and steel pipe is inserted into ground 500 mm deep. Greenhouse top is covered with transparent plastic film, to prevent rainwater interference. The airtight gauze net is used to enclose greenhouse to prevent pests. The greenhouse sets four fans for ventilation and heat dissipation, to make sure that the vegetables do not wither due to high temperature. The experimental equipment is shown in Fig. 1.

2.1.2 Vegetable pot. The vegetable pot is made of plastic material, with the volume of 20 L. Its diameter is 300 mm and its height is 400 mm, with a tray below, as shown in Fig. 2.

2.1.3 Experimental soil. 3.5 t of soil (sandy loam) with good texture is selected nearby, and the soil is dried in the air outdoors. After removing the grass roots, gravel and other impurities, the soil is sifted with a 20-mesh sieve. 500 kg of decomposed manure and right amount of inorganic fertilizers are mixed with the soil to form the experimental soil. After loading the tray with soil, the soil is fully watered, and the film is covered to preserve moisture. After the water never drips, it is placed in the vegetable

Received: March 26, 2015 Accepted: May 17, 2015

Supported by Key Science and Technology Project of Tianhe District, Guangzhou City, Guangdong Province (201205YG088).

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greenhouse. At the same time, some soil samples are dried and weighed for testing and calculating the soil pH and heavy metals, as shown in Table 1. Except cadmium, all heavy metals can basi-

cally meet the secondary standard of Soil Environmental Quality Standards (GB15618 – 1995), as shown in Table 2.

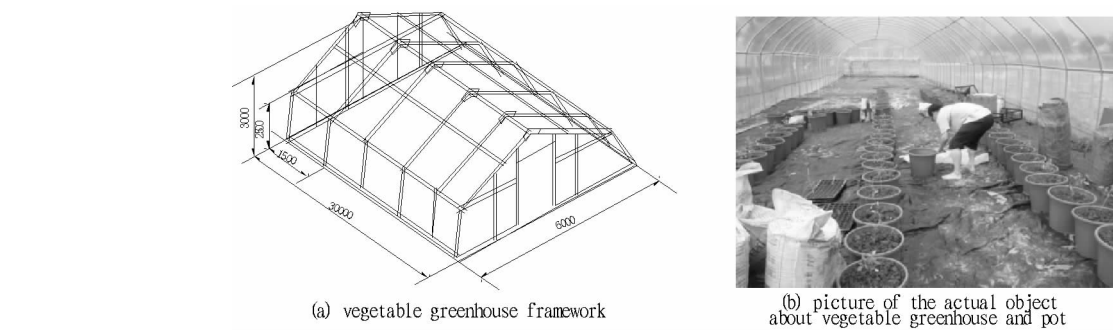


Fig.1 Experimental equipment

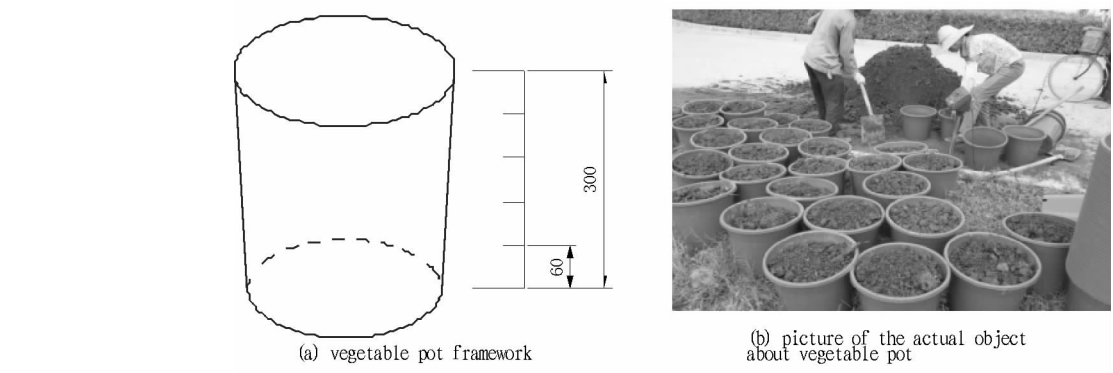


Fig.2 Vegetable pot

| Table 1 Heavy metal content | | | | | | | | | Unit: mg/kg |
|-----------------------------|-----|---------|---------|--------|--------|--------|-------|-------|-------------|
| Detection indicators | pH | Mercury | Arsenic | Chrome | Copper | Nickel | Lead | Zinc | Cadmium |
| Value | 7.5 | 0.23 | 12.43 | 26.21 | 28.79 | 11.83 | 22.03 | 69.95 | 1.40 |

| Table 2 The limit values of soil environmental quality | | | | | | Unit: mg/kg |
|---|--------------------|--------------|---------------|---------|-------------|-------------|
| Items | First level | Second level | | | Third level | |
| | Natural background | pH <6.5 | 6.5 < pH <7.5 | pH >7.5 | pH >6.5 | |
| Mercury | 0.15 | 0.30 | 0.50 | 1.0 | 1.5 | |
| Arsenic | 15 | 40 | 30 | 25 | 40 | |
| Copper | 35 | 50 | 100 | 100 | 400 | |
| Lead | 35 | 250 | 300 | 350 | 500 | |
| Chrome | 90 | 150 | 200 | 250 | 300 | |
| Zinc | 100 | 200 | 250 | 300 | 500 | |
| Nickel | 40 | 0 | 50 | 60 | 200 | |
| Cadmium | 0.20 | 0.30 | 0.30 | 0.60 | 1.0 | |

2.1.4 Experimental water. We select the sewage of Guangzhou Shijing Sewage Treatment Plant, reclaimed water and tap water as

the experimental irrigation water. The water quality is shown in Table 3.

| Table 3 Experimental water indicators | | | | | | | | | Unit: mg/L |
|---------------------------------------|-----|---------|---------|---------|---------|---------|---------|--------|------------|
| Detection indicators | pH | Mercury | Arsenic | Chrome | Copper | Nickel | Lead | Zinc | Cadmium |
| Tap water | 7.1 | <0.0010 | <0.0010 | <0.0100 | <0.0100 | <0.0100 | <0.0500 | 0.0160 | <0.0030 |
| Reclaimed water | 7.2 | 0.0001 | 0.0043 | 0.0160 | 0.0200 | 0.0210 | <0.0500 | 0.0360 | <0.0030 |
| Sewage | 7.5 | 0.0005 | 0.0056 | 0.0390 | 0.3180 | 0.0650 | <0.0500 | 0.2990 | <0.0030 |

2.2 Experimental methods The experiment sets two repeated irrigation plots, and we select the experimental pots in Plot I as

the research object (Table 4). The distribution of experimental plot is shown in Fig. 3. Five pots of vegetables are planted in each group, and in vegetable growth process, it is watered once a day at 8:00 and 16:00, each about 1 L. The experiment started from September 1, 2013 to December 31, 2013. The samples are taken

in the first day of each month. 100 g of mixed samples are taken from the pot irrigated with tap water; 100 g of samples 0 mm, 60 mm, 120 mm, 180 mm and 240 mm deep in the pots irrigated by reclaimed water and sewage, respectively.

Table 4 Experimental design and arrangement of irrigation water for vegetables

| Plot I | | | Plot II | | |
|--------------------------|--------------------------|--------------------------|---------------------------|---------------------------|---------------------------|
| Group I; tap water | Group I; reclaimed water | Group I; sewage | Group II; sewage | Group II; reclaimed water | Group II; tap water |
| Group I; reclaimed water | Group I; sewage | Group I; tap water | Group II; tap water | Group II; sewage | Group II; reclaimed water |
| Group I; sewage | Group I; tap water | Group I; reclaimed water | Group II; reclaimed water | Group II; tap water | Group II; sewage |



Fig.3 The distribution of experimental plot

2.3 Sample treatment

The soil samples are first placed in

the laboratory to be aired naturally, and then dried in an oven at 105 °C. After sample grinding, digestion and other pretreatment processes, the samples are detected and analyzed. Chromium, cadmium, copper, zinc, nickel and lead are detected using atomic absorption spectrometry while mercury and arsenic are detected using atomic fluorescence spectrophotometry.

3 Results and analysis

By experiment, it is found that the heavy metal content is low in the vegetable soil irrigated by reclaimed water while the heavy metal content is high in the vegetable soil irrigated by sewage. The experimental data are shown in Table 5.

Table 5 The content of heavy metal in the experimental vegetable soil

| Date (month. day) | Sample name | Soil depth mm | Mercury mg/kg | Arsenic mg/kg | Chrome mg/kg | Copper mg/kg | Nickel mg/kg | Lead mg/kg | Zinc mg/kg | Cadmium mg/kg |
|----------------------|-----------------|---------------------|------------------|------------------|-----------------|-----------------|-----------------|---------------|---------------|------------------|
| 9.1 | | Original sample | 0.23 | 12.43 | 26.21 | 28.79 | 11.83 | 22.03 | 69.95 | 1.40 |
| 10.1 | Tap water | Average sample | 0.23 | 12.43 | 33.63 | 17.38 | 14.51 | 18.51 | 62.50 | 1.44 |
| 11.1 | Tap water | Average sample | 0.30 | 12.58 | 35.21 | 17.22 | 14.00 | 18.58 | 60.60 | 1.48 |
| 12.1 | Tap water | Average sample | 0.30 | 13.41 | 35.97 | 13.62 | 14.90 | 20.92 | 59.62 | 1.54 |
| 10.1 | Reclaimed water | 0 | 0.14 | 10.30 | 32.96 | 23.37 | 15.03 | 22.93 | 74.53 | 1.84 |
| 11.1 | Reclaimed water | 0 | 0.50 | 12.11 | 44.93 | 14.85 | 28.60 | 24.55 | 59.46 | 1.77 |
| 12.1 | Reclaimed water | 0 | 0.86 | 60.68 | 42.74 | 24.88 | 28.61 | 25.71 | 82.77 | 2.10 |
| 10.1 | Reclaimed water | 60 | 0.28 | 10.80 | 48.40 | 25.32 | 39.87 | 25.75 | 80.30 | 1.97 |
| 11.1 | Reclaimed water | 60 | 0.18 | 12.19 | 34.90 | 25.12 | 24.28 | 26.08 | 78.06 | 1.97 |
| 12.1 | Reclaimed water | 60 | 0.24 | 11.22 | 35.04 | 21.59 | 24.86 | 23.26 | 68.95 | 1.94 |
| 10.1 | Reclaimed water | 120 | 0.38 | 12.69 | 42.70 | 19.99 | 25.83 | 21.56 | 80.17 | 1.46 |
| 11.1 | Reclaimed water | 120 | 0.16 | 13.03 | 41.42 | 19.83 | 26.83 | 19.18 | 65.74 | 1.46 |
| 12.1 | Reclaimed water | 120 | 0.17 | 10.47 | 42.79 | 21.76 | 27.07 | 17.94 | 69.84 | 1.38 |
| 10.1 | Reclaimed water | 180 | 1.14 | 10.30 | 43.07 | 22.29 | 27.43 | 19.70 | 88.62 | 1.63 |
| 11.1 | Reclaimed water | 180 | 0.33 | 10.62 | 33.27 | 24.12 | 23.30 | 18.28 | 75.86 | 1.50 |
| 12.1 | Reclaimed water | 180 | 0.40 | 11.40 | 32.10 | 20.61 | 22.72 | 21.49 | 69.27 | 1.46 |
| 10.1 | Reclaimed water | 240 | 0.23 | 15.30 | 33.50 | 20.30 | 23.47 | 19.86 | 68.75 | 1.37 |
| 11.1 | Reclaimed water | 240 | 0.55 | 10.39 | 38.81 | 24.80 | 32.40 | 23.42 | 67.73 | 1.59 |
| 12.1 | Reclaimed water | 240 | 0.32 | 11.72 | 39.38 | 14.26 | 26.51 | 19.80 | 60.83 | 1.47 |
| 10.1 | Sewage | 0 | 0.62 | 10.60 | 30.16 | 34.38 | 13.90 | 15.90 | 90.78 | 1.69 |
| 11.1 | Sewage | 0 | 0.46 | 12.52 | 50.99 | 18.27 | 22.50 | 17.69 | 61.36 | 1.44 |
| 12.1 | Sewage | 0 | 0.41 | 12.84 | 57.77 | 24.69 | 23.02 | 20.94 | 74.15 | 1.97 |
| 10.1 | Sewage | 60 | 0.25 | 7.70 | 62.34 | 26.71 | 28.33 | 23.86 | 77.66 | 1.98 |
| 11.1 | Sewage | 60 | 0.18 | 11.92 | 50.69 | 27.47 | 23.40 | 23.03 | 76.22 | 2.10 |
| 12.1 | Sewage | 60 | 0.19 | 12.48 | 88.84 | 18.86 | 38.22 | 18.43 | 69.51 | 1.47 |
| 10.1 | Sewage | 120 | 0.36 | 12.76 | 37.97 | 16.31 | 17.24 | 23.75 | 62.63 | 1.56 |
| 11.1 | Sewage | 120 | 0.25 | 15.71 | 38.54 | 10.38 | 18.13 | 14.69 | 55.57 | 1.82 |
| 12.1 | Sewage | 120 | 0.92 | 12.23 | 77.92 | 22.36 | 34.17 | 15.54 | 70.92 | 1.50 |
| 10.1 | Sewage | 180 | 0.57 | 12.02 | 33.47 | 23.98 | 15.06 | 24.95 | 69.38 | 1.48 |
| 11.1 | Sewage | 180 | 0.14 | 12.84 | 48.31 | 17.83 | 22.17 | 15.48 | 64.43 | 1.51 |
| 12.1 | Sewage | 180 | 0.47 | 14.09 | 72.21 | 19.08 | 31.63 | 17.36 | 72.44 | 1.80 |
| 10.1 | Sewage | 240 | 0.34 | 12.38 | 34.76 | 23.02 | 15.18 | 16.96 | 72.37 | 1.53 |
| 11.1 | Sewage | 240 | 0.71 | 11.91 | 35.98 | 21.47 | 15.28 | 20.66 | 72.95 | 1.51 |
| 12.1 | Sewage | 240 | 0.36 | 12.66 | 62.31 | 18.16 | 28.07 | 18.74 | 66.57 | 1.42 |

Based on the value of soils irrigated with tap water, we choose the representative chrome and nickel as the research object for data analysis, and get the average values of chrome and nickel in 0 mm, 60 mm, 120 mm, 180 mm and 240 mm deep soils in the same time period, with the time as the abscissa and chromium and nickel concentrations as vertical axis (Fig. 4a). From the curve in Fig. 4a, it is found that the content of heavy metals in the soil irrigated with sewage gradually increases over time, and the increasing trend basically meets the increment mode of simple cubic equation. The content of heavy metals in the soil irrigated with reclaimed water slightly increases, and the accumulation of heavy metals in soil basically meets the logarithmic function trend, and the content of heavy metals basically tends to be at a stable level over time, indicating that the accumulation of heavy metals in soil is a dynamic balance process, because the content of heavy metals in reclaimed water is low, and in the process of using reclaimed water to irrigate soil, part of accumulated heavy metals seep with water flow. The concentration of heavy metals in sewage is high, the accumulated heavy metals are more than the heavy metals seeping out over time, and the heavy metal content shows an increasing trend. The content of heavy metals in the soil in Fig. 4a is significantly lower than in Fig. 4b. Based on Table 1 and Table 3, it can be found that the content of heavy metals in soil is closely related to the background value of the original soil, and the irrigation water has a small impact. The content of nickel in the soil irrigated by reclaimed water and sewage is lower than 40 mg/kg, and the content of heavy metals in the soil irrigated by reclaimed water tends to be at a stable level.

The samples at different depths are taken and the concentration of chrome and nickel is detected. With the depth as abscissa and concentration as vertical axis, we get Fig. 5. From the curves in the figure, it is found that in the vegetable soil irrigated with reclaimed water, the heavy metal content shows a substantially horizontal trend, and with the change of time and depth, the changes in the concentration of heavy metals become small. The content of heavy metal in reclaimed water is low, and the accumulation in soil follows the laws of dynamic equilibrium; the accumulation

amount is basically equal to desorption amount, and the concentration value is close to the background value of soil, so the heavy metal content is higher than the original level and always shows a steady state. The soil irrigated with sewage shows a quadratic polynomial decreasing trend. The heavy metal content presents the maximum value at 60 mm, and then the concentration gradually decreases with the increasing depth, indicating that in the soil surface, the amount of heavy metals absorbed by soil is far greater than the desorption amount, but with the increasing depth of the soil, the heavy metal content of the water decreases, leading to decreasing adsorption amount and increasing desorption amount, so the content of heavy metals in soil gradually decreases. As can be seen from the figure, the heavy metal content gradually increases over time, which is consistent with the case in Fig. 4. LEC risk assessment method is used for the risk assessment on the soil irrigated by reclaimed water. The sewage treatment plant detects and analyzes the reclaimed water every day to ensure that the water meets the vegetable irrigation standards, and through the experiments, it is found that the content of heavy metal in the soil irrigated with reclaimed water is not increased obviously, and remains stable to meet the standard requirements of vegetable cultivation, so the possibility of accidents is "very unlikely", and L value is 0.5. After using the reclaimed water for the irrigation of vegetables, whether the heavy metal in soil is adsorbed by vegetables is to be studied in the further experiment. Assuming the vegetables come into contact with irrigation water, and human body is indirectly put in the risk environment through the consumption of vegetables; human will eat vegetables every day, so the frequency of exposure to the risk environment is at the highest level "continuous exposure", and E value is 10. The content of heavy metal in reclaimed water is low, and the heavy metal accumulated in soil is also low, meeting the needs of growing vegetables, so there is basically no risk of harm, and the consequence of an accident is "minor injuries", with C value of 1. The risk score D ($D = L \times E \times C$) is calculated at 5, less than 20, a little dangerous, so the soil irrigated with reclaimed water is at the level of tolerable risk (safe, no need to take action).

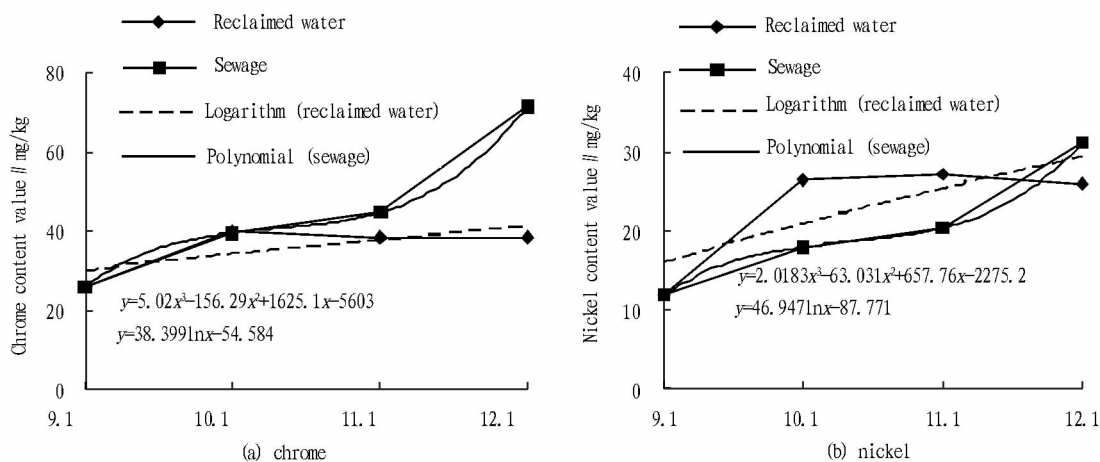


Fig. 4 The accumulation of chrome and nickel in the vegetable soil

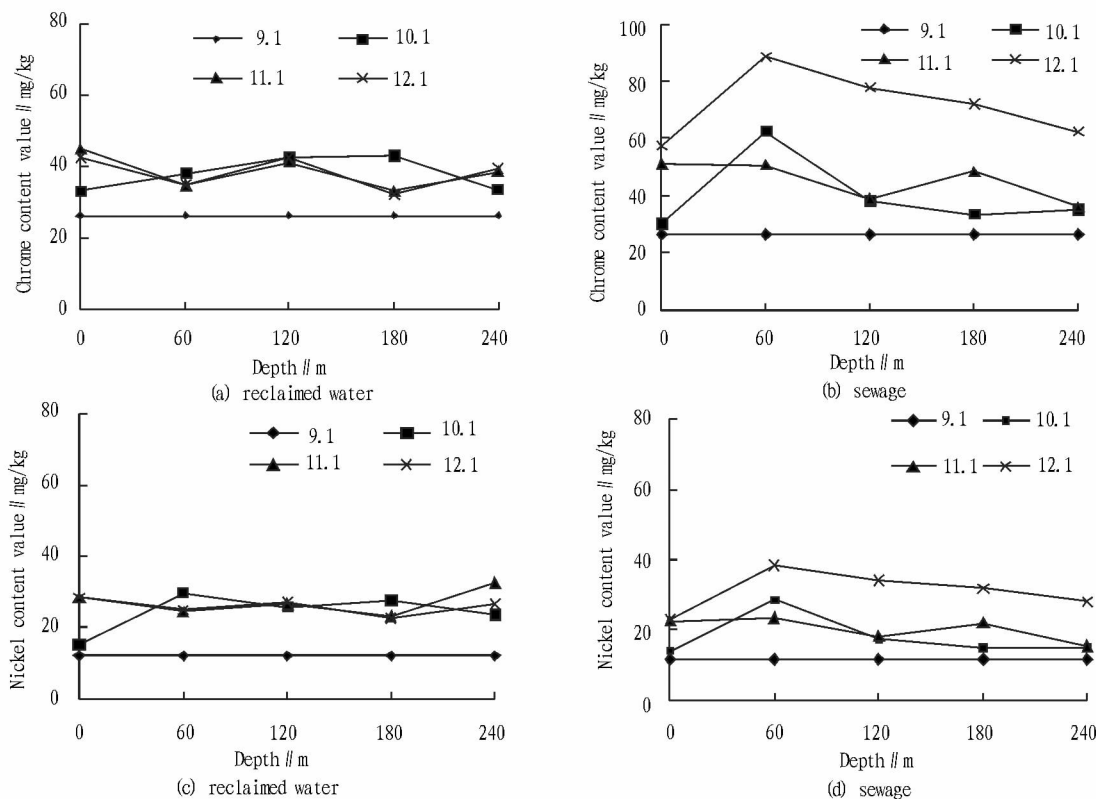


Fig. 5 The changes in the concentration of heavy metals in the vegetable soil irrigated by reclaimed water or sewage with soil depth

4 Conclusions and discussions

From the experimental study, it is found that compared with the vegetable soil irrigated with sewage, the content of heavy metal in the soil irrigated with reclaimed water basically meets the secondary standard of *Soil Environmental Quality Standards* (GB15618 – 1995). There is accumulation of heavy metal in soil during irrigation, but over time, the heavy metals accumulated in soil gradually realize desorption and show dynamic balance, and the topsoil for vegetable cultivation is in full compliance with the requirements of environmental quality. LEC risk assessment method is used for the risk assessment on the soil irrigated by reclaimed water, and it is at the level of tolerable risk (safe, no need to take action). The study clears the migration of heavy metal in soil, the risk of contamination of shallow groundwater and heavy metal accumulation in soil during the irrigation of vegetables with reclaimed water, improves the theoretical researches in areas of reclaimed water, and further corroborates the conclusion of Lahaam *et al.* that the reclaimed water is basically safe to be irrigation water. Based on the actual situation of water pollution in Guangzhou City, this study aims to provide the most effective irrigation water for the planting of vegetables in Guangzhou City. The cadmium in the used soil slightly exceeds the secondary standard of *Soil Environmental Quality Standards* (GB15618 – 1995), but it has little effect on the studies of migration of migration in soil irrigated with

reclaimed water. Findings confirm that the reclaimed water is basically safe to be irrigation water, but there is a need to refine the reclaimed water use practices to ensure the safety of reclaimed water in the use process. Before the use of reclaimed water, it is necessary to test the reclaimed water quality to ensure that the water reaches the standard and further study incidental and uncertain risks.

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detrimental to health, and will consider mixed application of chemical and organic fertilizers.

Table 4 Model regression results

| Variables | Regression coefficient | t value |
|-------------------|------------------------|---------|
| Constant | 337.752*** | 3.49 |
| Char1 | -130.194** | 2.28 |
| Char2 | -112.326*** | -5.26 |
| Oper1 | 0.001 | -0.31 |
| Oper 2 | -101.425** | -2.21 |
| Oper 3 | 64.053 | 0.88 |
| Oper 4 | 152.515*** | 3.01 |
| Oper 5 | -14.523 | -0.50 |
| CTC | -9.622 | -0.26 |
| SFA1 | 17.878 | 0.32 |
| SFA2 | 3.325 | 0.10 |
| SFA3 | -59.477 | -1.19 |
| Number of samples | 340 | |
| A-R2 | 0.12 | |
| F value | 4.13 | |

Note: ***, ** and * signify that variable is significant at 1%, 5% and 10% respectively.

3.3.4 Scientific fertilizer application ability. All three variables of scientific fertilizer application ability, SFA1, SFA2 and SFA3, fail to pass the significance test. This is possibly because farmers seldom participate in agricultural technological training and extension of scientific fertilizer application technologies, and professional level of agricultural technological personnel is to be improved.

4 Conclusions

Improper use of chemical fertilizers is an essential reason for diffused pollution of agriculture. Therefore, finding out influence factors of farmers in application of chemical fertilizers will play a significant role in controlling the diffused pollution of agriculture. Based on survey data of 340 farmers in Chongqing, we made empirical analysis on influence factors of farmers' application of chemical fertilizers, including basic characteristics of farmer householder, farmer family and production characteristics, agricultural product trading characteristics, and farmers' scientific fertilizer application ability. Results show that application of chemi-

cal fertilizers is negatively correlated with age and education level of farmer householder, while women labors are more likely to apply more chemical fertilizers. Besides, low soil fertility will lead to more application of chemical fertilizers. Family annual income, proportion of agricultural production population, commodity trading characteristics, and whether organic fertilizers used or not fail to pass the significance test. Finally, the influence of scientific fertilizer application ability on farmers' application of chemical fertilizers is not significant.

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