Heavy metal pollution characteristics and assessment of environmental quality and safety of facility agriculture soil in Shouguang

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Abstract. Taking Shouguang Facility agriculture in Weifang City as the research object, heavy metals and other pollutants in new and old facility agriculture soil were investigated and analyzed from 2017 to 2019. The content of heavy metals in soil was analyzed, and the environmental quality of heavy metals was evaluated by single-factor pollution index method and Nemerow index method. The correlation between heavy metal content in surface soil and cultivation years, vertical distribution of heavy metal content in soil, frequency distribution of heavy metal in surface soil, and correlation between heavy metals in surface soil and soil nutrients were analyzed. The results showed that the average contents of the eight heavy metals were Cd 0.23 mg•kg⁻¹, Hg 0.05 mg•kg⁻¹, As 8.09 mg•kg⁻¹, Pb 23.31 mg•kg⁻¹, Cr 70.81 mg•kg⁻¹, Cu 38.54 mg•kg⁻¹, Ni 25.87 mg•kg⁻¹ and Zn 147.52 mg•kg⁻¹. Aside from Pb and Ni, the average content of all other heavy metals in the surface soil of facility agriculture was higher than the average of the control and the geochemical background level of soil in Weifang. Compared with the Environmental Quality Evaluation Standard for Farmland of Greenhouse Vegetables Production (HJ333-2006) and the Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (trial) (GB15618-2018), the average content of heavy metals met the standards, with Cd, Hg and Zn content in some samples exceeding the standards. Most surface soil environmental quality is clean or relatively clean, but there is also part of the soil has reached the level of mild or moderate pollution. The content of heavy metals in the soil of the old facility agriculture (cultivation years > 10) was higher than that of the new facility agriculture (cultivation years ≤ 10), and the content of heavy metals in the surface soil (0~20cm) was higher than that in the deep soil. The quality distribution of heavy metals in the surface soil is affected by exogenous heavy metals, among which Hg and Cd have the largest skewness coefficient and are most affected by exogenous. There was significant positive correlation between heavy metals in surface soil and available phosphorus, hydrolytic nitrogen, available potassium and organic matter. The results indicated that the accumulation of heavy metals in soil was related to the application of organic fertilizer, phosphate fertilizer and compound fertilizer.

1. Introduction
Along with the expansion of cultivated area and the increase in cultivation years of facility agriculture in China, the microecological environment within facility agriculture has gone through significant changes; moreover, the long-term and heavy use of pesticide, chemical fertilizers and organic fertilizers has resulted in secondary salinization, acidification, microflora damage and a series of other
problems of quality degradation and continuous cropping obstacles in facility agriculture soil[1-2]. In the next place, long-term and heavy application of organic fertilizers, chemical fertilizers, pesticide, etc. has led to the increasingly severe problem of nitrogen, phosphorus and other agricultural non-point source pollution. These existing problems have resulted in greater risks of environmental pollution and for health and safety, and have bottlenecked the sustainable development of facility agriculture [3].

According to related research, facility agriculture soil featured the heavy metal migration capacity better than general uncovered soil [4], heavy metals in which, instead of being degraded by microorganisms, eventually entered the human body via the food chain and endangered human health [5-7]. In addition to heavy metals from parent materials, application of pesticide and chemical and organic fertilizers containing heavy metals would also lead to excessive lead, mercury, cadmium, arsenic and other heavy metals in the facility agriculture soil [8-13]. As reported by Li Jianyun et al. [14-15], the content of heavy metals such as copper, zinc and lead increased to a certain extent as the cultivation years increased, while cadmium content showing no obvious pattern; however, all these heavy metals showed higher content than in farmland. Li Decheng et al. [16] noticed that the content of most heavy metals in facility agriculture soil increased as the service life of the greenhouse extended, yet no excessive level of heavy metal pollution was detected. The harm of heavy metals is not only dependent on their total amount, but also closely related to their occurrence mode in soil. Heavy fertilizer application in facility cultivation was an important influencing factor of the physical and chemical properties of soil and moreover, the available content of heavy metals in soil, and a main path leading to agricultural non-point source pollution [17]. Foreign research focused more on the film material properties of greenhouse facilities, the efficiency and balance of greenhouse heating [18-19], growth and yield of crops under protected cultivation [20-21], utilization of water for irrigation and the effect of pest control [22], etc., while mainly adopting soilless cultured flowers (not to enter the food chain) as research subjects. Therefore, related research on heavy metals in facility culture was rarely found.

Shouguang is the “City of Vegetables in China”, with large-scale and various vegetable cultivation. After years’ development, Shouguang facility culture has become the local pillar and characteristic industry. However, few systematic reports were available on the characteristics of pollution, temporal and spatial evolution, and environmental quality and safety in its facility agriculture. In this research, with the famous vegetable base Shouguang of China as the subject, the characteristics of heavy metal pollutants in old and new facility agriculture soil in 2017~2019 were discussed. Statistical characteristics of heavy metals in surface soil were analyzed, and the environmental quality of heavy metals evaluated by single-factor pollution index and Nemerow index. Meanwhile, the correlation between heavy metal content in surface soil and cultivation years, the vertical distribution of heavy mental content in soil, the frequency distribution of heavy metals in surface soil, and the correlation between heavy metal content in surface soil and soil nutrients analyzed statistically. In this way, sources of heavy metals in facility agriculture soil were revealed, with a view to providing a scientific basis for the control of heavy metal risks in soil and the safety of agricultural products under facility agriculture.

2. Materials and Methods

2.1. Overview of the research area
Shouguang City (118°32′~119°10′E, 36°41′~37°19′N) is located in Weifang City, China, in the north-central Shandong Peninsula, on the southwest coast of Laizhou Bay, covering a total area of 2180km², all plain (Figure 1). Shouguang is the place of origin of China’s winter-heated greenhouses, the largest vegetable production base nationwide and the well-known “City of Vegetables in China”, with over 53,000 hectares of vegetable cultivation. The place is of semi-humid warm temperature climate, with abundant sunshine and four distinct seasons. Annual mean temperature there is about 12.40℃, average
annual rainfall about 608mm, and frost-free season around 195d; the main soil types are cinnamon soil and meadow cinnamon soil, soil pH being neutral or weak alkaline.

2.2. Collection of soil samples and test methods
Soil samples of facility agriculture in Shouguang were collected in 2017~2019. Figure 1 is the map of soil sampling sites.

![Figure 1. The location of sample distribution.](image)

In this research, based on the actual cultivated area, types, quantity and scale and cultivation years of facility agriculture in the villages and towns, samples of soil were collected from representative facility agriculture there, and samples from fields, bare soil, etc. receiving smaller disturbance of human activities collected as control. 93 soil samples were collected in total, of which 65 were from the surface soil 0~20cm deep under facility agriculture, 8 from the surface soil 0~20cm deep of the control sites (Table 1). In addition, to compare the heavy metal content in different layers of soil, this research also adopted stratified continuous sampling in 0~20cm, 20~40cm, 40~60cm, 60~80cm and 80~100cm (sampling sites 1, 6, 12, 18 and 48), totaling 20 deep soil samples.
### Table 1. The sampling conditions of facility agriculture.

| Site          | Number of sampling sites | Vegetables                                      |
|---------------|--------------------------|-------------------------------------------------|
|               | Total                    | Cultivation years > 10 | Cultivation years ≤ 10 |
| Daotian Town  | 10                       | 4                   | 6                      |
|               |                          | Haimi melons, tomatoes, small tomatoes, cantaloupes, peppers, cucumber, chilies, cauliflower, loofahs, kidney beans |
| Luocheng St.  | 14                       | 10                  | 4                      |
| Hou Town      | 5                        | 5                   | 0                      |
| Hualong Town  | 8                        | 8                   | 0                      |
| Wenjia St.    | 6                        | 2                   | 4                      |
| Tianliu Town  | 4                        | 1                   | 3                      |
| Jitai Town    | 9                        | 5                   | 4                      |
| Sunjiaji St.  | 9                        | 5                   | 4                      |
|               |                          | peppers, cucumbers, loofahs                      |
|               |                          | peppars, cucumbers, yam                         |
|               |                          | kidney beans, cucumbers, tomatoes                |
|               |                          | pumpkins, eggplants                              |
|               |                          | bitter gourd, cabbage, spinach, broccoli         |

Each soil sample was the mixture of soil from 5 points distributed in “S” shape at the same sampling site. Samples collected were ground in an agate mortar after air-dry, then sieved with a 100 mesh (0.15mm) nylon sieve to measure the content of heavy metals, hydrolysable nitrogen, available phosphorus, available potassium and organic matter in soil. Total cadmium (Cd) and total lead (Pb) in soil were measured by graphite furnace atomic absorption spectrometry (GFAAS); total chromium (Cr), total copper (Cu), total zinc (Zn) and total nickel (Ni) were measured by inductively coupled plasma optical emission spectrophotometry (ICP-OES); total hydrargyrum (Hg) and total arsenic (As) were measured by atomic fluorescence spectrometry (AFS); hydrolysable nitrogen in soil was measured by alkali N-proliferation method; available phosphorus was measured by sodium bicarbonate extraction-Mo-Sb anti-spectrophotometer method; available potassium was measured by ammonium acetate extraction-flame photometry, and organic matter was measured by potassium dichromate oxidation-external heating method; pH was measured by potentiometric method.

#### 2.3. Data processing

Statistics of heavy metals in soil, analysis of the frequency distribution of heavy metals, and the correlation analysis were all completed with SPSS 26.0, and other data analysis by Excel 2007. The map of soil sampling sites was drawn by ArcGis 10.2, and other figures by Origin 9.1.

#### 2.4. Evaluation methods and standards of environmental quality of heavy metals in soil

There were many methods for evaluation of heavy metal pollution in soil [23-24]. In this research, referring to the evaluation methods and standards in Environmental Quality Evaluation Standard for Farmland of Greenhouse Vegetables Production (HJ333-2006, National standard) and Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (trial) (GB15618-2018, National standard), the method of single-factor pollution index and Nemerow index method were adopted to evaluate the environmental quality of heavy metals. See Table 2 and Table 3 for the evaluation standards.

### Table 2. Environmental Quality Evaluation Standard for Farmland of Greenhouse Vegetables Production- Limits of soil environmental quality evaluation index.

| pH | Cd  | Hg  | As  | Pb  | Cr  | Cu  | Ni  | Zn  |
|----|-----|-----|-----|-----|-----|-----|-----|-----|
|    | mg·kg⁻¹ |     |     |     |     |     |     |     |
| <6.5 | 0.30 | 0.25 | 30  | 50  | 150 | 50  | 40  | 200 |
| 6.5-7.5 | 0.30 | 0.30 | 25  | 50  | 200 | 100 | 50  | 250 |
| >7.5 | 0.40 | 0.35 | 20  | 50  | 250 | 100 | 60  | 300 |
Table 3. Risk control standard for soil contamination of agriculture land—Risk screening values for soil contamination of agricultural land.

| pH | Cd | Hg | As | Pb | Cr | Cu | Ni | Zn |
|----|----|----|----|----|----|----|----|----|
| 5.5<pH≤6.5 | 0.3 | 1.8 | 40 | 90 | 150 | 50 | 70 | 200 |
| 6.5<pH≤7.5 | 0.3 | 2.4 | 30 | 120 | 200 | 100 | 100 | 250 |
| >7.5 | 0.6 | 3.4 | 25 | 170 | 250 | 100 | 100 | 250 |

The computing formula of single-factor pollution index method is:

\[ P_i = \frac{C_i}{S_i} \]  
(1)

Where \( P_i \) is the single-factor pollution index of heavy metal element \( i \) in soil, \( C_i \) is the measured content of heavy metal \( i \) in soil, and \( S_i \) is the evaluation standard of heavy metal element \( i \).

Comprehensive evaluation of soil environmental quality adopted Nemerow formula to work out the comprehensive pollution index [25], whose results not only considered the average level of pollution of different pollutants, but also reflected environmental damage caused by the most polluting pollutant [26]. The computing formula is:

\[ P = \sqrt{\frac{P_{i\text{avg}}^2 + P_{i\text{max}}^2}{2}} \]  
(2)

Where \( P \) is the Nemerow index, \( P_{i\text{avg}} \) is arithmetic average of the pollution indexes, and \( P_{i\text{max}} \) is the maximum single-factor pollution index of the pollutants.

Table 4 is the evaluation standards of single-factor pollution index and Nemerow index.

| Single-factor pollution index /Nemerow index | level                  |
|--------------------------------------------|------------------------|
| \( P_i \leq 0.7 \) / \( P \leq 0.7 \)       | Clean                  |
| \( 0.7 < P_i \leq 1.0 \) / \( 0.7 < P \leq 1.0 \) | Relatively clean       |
| \( 1.0 < P_i \leq 2.0 \) / \( 1.0 < P \leq 2.0 \) | Mild pollution         |
| \( 2.0 < P_i \leq 3.0 \) / \( 2.0 < P \leq 3.0 \) | Moderate pollution     |
| \( P_i > 3.0 \) / \( P > 3.0 \)             | Serious pollution      |

Standard-exceeding rate is the ratio (in percentage) of total number of standard-exceeding samples to the total number of samples surveyed. The computing formula is:

\[ \text{Standard-exceeding Rate} = \left( \frac{\text{Total Number of Standard-exceeding Samples}}{\text{Total Number of Samples Surveyed}} \right) \times 100\% \]  
(3)

3. Results and discussion

3.1. Statistical analysis of heavy metal content in surface soil (0~20cm)

Table 5 is the statistical analysis of heavy metal content in surface soil. Surface soil pH of facility agriculture was 6.19~8.38, of which 78.46% points fell within 6.5~7.5, and 20% above 7.5. Therefore, the soil there was neutral to alkaline. The average content of eight heavy metals was: Cd 0.23mg·kg⁻¹, Hg 0.05mg·kg⁻¹, As 8.09mg·kg⁻¹, Pb 23.31mg·kg⁻¹, Cr 70.81mg·kg⁻¹, Cu 38.54mg·kg⁻¹, Ni 25.87mg·kg⁻¹ and Zn 147.52mg·kg⁻¹. According to the coefficient of variation (CV), different facility agriculture soil showed significant differences in heavy metal content, CV being 10%–97.30%. Among others, CV of Hg and Cd reached 97.30% and 91.30% respectively, and that of both Cu and
Zn above 40%; Pb and Cr showed lower CV. CV of different elements in soil was in turn: Hg > Cd > Cu > Zn > Ni > Cr > As > Pb.

Aside from Pb and Ni, the average content of all other heavy metals in the surface soil of facility agriculture was higher than the average of the control and the geochemical background level of soil in Weifang. Compared with the average of the control, the content of Cd, Hg, Cu and Zn in facility agriculture soil showed the biggest increase, by 0.77, 0.67, 0.72 and 1.27 times respectively; the content of As, Pb, Cr and Ni remained basically equal to the average of the control. Compared with the geochemical background level of soil in Weifang, the content of Cd, Hg, Cu and Zn in facility agriculture soil showed the biggest increase, by 1.02, 0.56, 0.82 and 1.52 times respectively; the content of As, Pb, Cr and Ni remained basically equal to the background level.

**Table 5.** Statistical characteristic values of heavy metals in surface soil of Shouguang facility agriculture (mg·kg⁻¹).

| Heavy metals | Number of samples | Content / mg·kg⁻¹ | Average /mg·kg⁻¹  | Standard deviation | Coefficient of Variation /% | Average of the control / mg·kg⁻¹ | Geochemical background level of soil in Weifang / mg·kg⁻¹ |
|--------------|-------------------|-------------------|-------------------|-------------------|-----------------------------|-----------------------------|--------------------------|
| Cd           | 65                | 0.06−1.14         | 0.23              | 0.21              | 91.30                       | 0.13                        | 0.114                    |
| Hg           | 65                | 0.02−0.37         | 0.05              | 0.05              | 97.30                       | 0.03                        | 0.032                    |
| As           | 65                | 4.41−13.44        | 8.09              | 1.76              | 21.70                       | 7.87                        | 7.8                      |
| Pb           | 65                | 17.30−30.10       | 23.31             | 2.32              | 10.00                       | 25.84                       | 25.98                    |
| Cr           | 65                | 37.59−118.50      | 70.81             | 17.11             | 24.20                       | 72.73                       | 65.3                     |
| Cu           | 65                | 16.87−99.14       | 38.54             | 17.26             | 44.80                       | 22.39                       | 21.20                    |
| Ni           | 65                | 0−48.66           | 25.87             | 9.72              | 37.60                       | 31.32                       | 26.9                     |
| Zn           | 65                | 54.04−353.60      | 147.52            | 62.76             | 42.50                       | 64.90                       | 58.5                     |

**3.2. Environmental quality and safety evaluation of heavy metals in surface soil**

By comparing the content of heavy metals with the average of the control and the geochemical background level of soil in Weifang, those of the highest standard-exceeding rates were Cd, Hg, Cu and Zn. Compared with Environmental Quality Evaluation Standard for Farmland of Greenhouse Vegetables Production (HJ333-2006) and Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (trial) (GB15618-2018), the average content of heavy metals met the standards, with Cd, Hg and Zn content in some samples exceeding the standards. Compared with HJ333-2006, the standard-exceeding rate of Cd, Hg, and Zn was 12.31%, 1.54% and 3.08% respectively; Compared with GB15618-2018, the standard-exceeding rate of Cd and Zn was 12.31% and 3.08% respectively (Table 6). The results basically agreed with the research of Li Shuhui et al[28].

**Table 6.** The situation of heavy metals in surface soil of facility agriculture exceeding the control and the relative standards.

| Heavy metals | Number of samples | Average of the control | Geographical background level of soil in Weifang | HJ333-2006 | GB15618-2018 |
|--------------|-------------------|------------------------|-------------------------------------------------|------------|--------------|
|              | Number of samples exceeding standard | Standard-exceeding rate (%) | Number of samples exceeding standard | Standard-exceeding rate (%) | Number of samples exceeding standard | Standard-exceeding rate (%) | Number of samples exceeding standard | Standard-exceeding rate (%) |
| Cd           | 65                | 43                     | 66.15                                           | 52         | 80.00        | 8                | 12.31                  | 8                        | 12.31                     |
| Hg           | 65                | 46                     | 70.77                                           | 43         | 66.15        | 1                | 1.54                   | 0                        | 0                         |
| As           | 65                | 28                     | 43.08                                           | 32         | 49.23        | 0                | 0                      | 0                        | 0                         |
Based on the evaluation standard of HJ333-2006 and GB15618-2018, the situation of heavy metal pollution in surface soil of facility agriculture was evaluated by single-factor pollution index ($P_i$) and Nemerow index ($P$) respectively (Table 7 and 8).

**Table 7.** Single-factor pollution index of heavy metals in surface soil of facility agriculture.

| Evaluation standard | Item | Cd  | Hg  | As  | Pb  | Cr  | Cu  | Ni  | Zn  |
|---------------------|------|-----|-----|-----|-----|-----|-----|-----|-----|
| HJ333-2006 $P_i$   | Average | 0.73 | 0.17 | 0.34 | 0.47 | 0.34 | 0.39 | 0.50 | 0.58 |
|                     | Standard deviation | 0.66 | 0.17 | 0.087 | 0.047 | 0.089 | 0.17 | 0.19 | 0.25 |
|                     | Minimum         | 0.15 | 0.05 | 0.18 | 0.35 | 0.19 | 0.19 | 0.00 | 0.21 |
|                     | Maximum         | 3.80 | 1.25 | 0.67 | 0.60 | 0.59 | 0.99 | 0.97 | 1.41 |
| GB15618-2018 $P_i$ | Average | 0.68 | 0.02 | 0.28 | 0.18 | 0.34 | 0.39 | 0.23 | 0.58 |
|                     | Standard deviation | 0.65 | 0.021 | 0.069 | 0.032 | 0.089 | 0.17 | 0.10 | 0.25 |
|                     | Minimum         | 0.10 | 0.005 | 0.15 | 0.10 | 0.19 | 0.19 | 0.00 | 0.21 |
|                     | Maximum         | 3.80 | 0.16 | 0.54 | 0.28 | 0.59 | 0.99 | 0.49 | 1.41 |

Based on HJ333-2006, the average of $P_i$ of Cd in surface soil of facility agriculture was 0.73, and the average of $P_i$ of other heavy metals smaller than 0.70; the average of $P_i$ in turn was: Cd$>$ Zn$>$ Ni$>$ Pb$>$ Cu$>$ Cr$>$ As$>$ Hg. The $P_i$ of As, Pb, Cr, Ni and Cu was smaller than 1, but the $P_i$ of Cd, Hg and Zn in some samples bigger than 1, of which the maximum $P_i$ of Cd was 3.80, way above the standard. Based on GB15618-2018, the average of $P_i$ of Cd in surface soil of facility agriculture was smaller than 0.70; the average of $P_i$ in turn was: Cd$>$ Zn$>$ Cu$>$ Cr$>$ As$>$ Ni$>$ Pb$>$ Hg. The $P_i$ of As, Pb, Cr, Ni, Cu and Hg was smaller than 1, but the $P_i$ of Cd and Zn in some samples bigger than 1. The above analysis indicated that, the level of Hg, Cd and Zn at some sample sites has reached mild to moderate pollution, and should be controlled in a timely manner.

**Table 8.** Nemerow index of heavy metals in surface soil of facility agriculture.

| Evaluation standard | Nemerow index Maximum | Minimum | Level | Number of samples | The proportion (%) |
|---------------------|-----------------------|--------|-------|------------------|--------------------|
| HJ333-2006          | 2.77                  | 0.35   | P≤0.7 | 50               | 76.92              |
|                     |                       |        | 0.7<P≤1.0 | 6   | 9.23              |
|                     |                       |        | 1.0<P≤2.0 | 7   | 10.77             |
|                     |                       |        | 2.0<P≤3.0 | 2   | 3.08              |
| GB15618-2018        | 2.76                  | 0.22   | P≤0.7 | 53               | 81.54              |
|                     |                       |        | 0.7<P≤1.0 | 5   | 7.69              |
|                     |                       |        | 1.0<P≤2.0 | 5   | 7.69              |
|                     |                       |        | 2.0<P≤3.0 | 2   | 3.08              |

Based on HJ333-2006, the Nemerow index of 86.15% surface soil of facility agriculture was smaller than 1, falling within clean or relatively clean levels; the Nemerow index of 10.77% soil was
1.0–2.0—mild pollution; the index of 3.08% soil was 2.0–3.0—moderate pollution. Benchmarked to GB15618-2018, the Nemerow index of 89.23% surface soil of facility agriculture was smaller than 1—clean or relatively clean; the index of 7.69% soil was between 1.0 and 2.0—mild pollution; the index of 3.08% soil was between 2.0 to 3.0—moderate pollution.

Therefore, the environmental quality of surface soil of facility agriculture in Shouguang was mostly clean or relatively clean, only part of the soil of mild or moderate level of pollution.

3.3. Analysis of correlation between heavy metal content in surface soil and cultivation years

To discuss the effect of cultivation years on the heavy metal content in facility agriculture soil in Shouguang, the soil samples were divided into new facility agriculture soil (cultivation years ≤ 10) and old facility agriculture soil (cultivation years > 10). The content of heavy metals was averaged and compared, as shown in Figure 2.

![Figure 2. Comparative analysis of heavy metals between old and new facility agricultures in Shouguang.](image)

The content of heavy metals in old facility agriculture soil was generally higher than that in new facility agriculture soil, of which Cd, Hg, Zn and Cu showed the biggest increase as the years of cultivation extended. Existing research has demonstrated that, the content of heavy metals in most facility agriculture soil increased along with the extension of service life[29]. Li Shuhui[28] and Qiao Debo[30] noticed that, as the years of cultivation extended, Cd, Cu and Zn showed the biggest increase, which basically agreed with this research.

3.4. Characteristics of the vertical distribution of heavy metal content in soil

To study characteristics of the vertical distribution of heavy metals in facility agriculture soil, in this research, soil samples were collected from different layers of soil of facility agriculture for analysis. The results are shown in Figure 3.
Figure 3. Distribution of heavy metal content in different soil depth of facility agriculture. 1, 6, 12, 18 and 48 are sampling sites. “New” stands for new facility agricultures; “Old” stands for old facility agricultures; “Control” stands for control sites.

It was generally recognized that heavy metals in soil were not easy to leach with water, and not degradable by soil microorganisms [31]. They were often strongly adsorbed and fixed in soil, most attached to the surface, not easy to migrate downwards. However, after being polluted by heavy metals, surface soil of high heavy metal content could move down by means of leaching [32]. The variation range of As, Pb, Cu, Ni, Zn, Cr, Hg and Cd in facility agriculture soil was 4.41~8.73 mg·kg\(^{-1}\), 19~25.37 mg·kg\(^{-1}\), 16.01~46.73 mg·kg\(^{-1}\), 0~47.93 mg·kg\(^{-1}\), 15.80~20.47 mg·kg\(^{-1}\), 51.67~115.97 mg·kg\(^{-1}\), 0.01~0.17 mg·kg\(^{-1}\) and 0.04~0.44 mg·kg\(^{-1}\) respectively. As and Pb content in soil increased first and then decreased from the surface (0~20cm) to deeper levels; the content of other heavy metals showed an overall declining trend, of which the variation of content at 60cm was greater, indicating that heavy metals in soil were mainly concentrated on the surface, some migrated deeper than 60cm and might further contaminating groundwater.

3.5 Analysis of the frequency distribution of heavy metals in surface soil

In natural environment, element content in soil was subject to the long-term and integrated effect of soil patent material composition, element migration and distribution in soil-forming process, biological factors, etc. The frequency distribution of element content usually agreed with the normal distribution or logarithmic normal distribution. However, affected by human disturbance, pollution and other factors, there could be skewed distribution because of element enrichment. The frequency histogram could display the characteristics of distribution of element content [27,33-35].

Figure 4 is the frequency distribution of heavy metal content in surface soil of facility agriculture in Shouguang. The frequency distribution of all 8 heavy metals in facility agriculture soil disagreed with standard normal distribution; however, the coefficient of skewness of Ni, Pb and As was smaller than 0.5, less affected by human disturbance. The coefficient of skewness of other heavy metals was bigger than 0.5—positive-skewed distribution; among others, Hg and Cd showed the highest skewness, the coefficient reaching 3 and above, with kurtosis coefficient bigger than 10. The positive-skewed distribution characteristics indicated certain accumulation of heavy metals in soil of the area, thus verifying the effect of exogenous heavy metals on the distribution of heavy metal content in surface soil of facility agriculture in Shouguang.
3.6. Analysis of the correlation between heavy metal content in surface soil and available phosphorus, hydrolysable nitrogen, available potassium and organic matter

Statistical analysis of heavy metal content in surface soil, calculation of Pearson correlation coefficient, and correlation analysis of heavy metal content indicated: a significant positive correlation between Hg and Cu and Zn (P<0.05), the coefficient of correlation (r) being 0.309 and 0.208 respectively, weak correlation; a significant positive correlation between Cd and Cr, Cu and Zn, of which the correlation with Cu and Zn was very significant (P<0.01), the coefficient of correlation between Cd and Cr being 0.307, weak correlation, and that between Cd and Cu and Cd and Zn being 0.568 and 0.426 respectively, moderate correlation; a very significant relationship between Cr and Cu, Ni and Zn (P<0.01), r being 0.360, 0.646 and 0.385 respectively, Cr and Ni being strongly correlated; a very significant correlation between Cu and Zn (P<0.01), r being 0.875, very strong correlation. Correlation of As and Pb with other elements was insignificant (Table 9).

Correlations between heavy metal content and available phosphorus, hydrolysable nitrogen, available potassium and organic matter revealed: a significant positive correlation between P and Cr, Cu and Zn, r being 0.266, 0.277 and 0.428 respectively; a significant positive correlation between N and As and Ni, r being 0.281 and 0.312 respectively; a significant positive correlation between K and Ni, r being 0.297; a significant positive correlation between organic matter and Cd, Cr, Cu and Zn, r being 0.262, 0.258, 0.425 and 0.566 respectively (Table 9).

The above correlation analysis indicated a certain level of synchronicity and homology among heavy metals and between heavy metals and available phosphorus, hydrolysable nitrogen, available potassium, organic matter, etc. in accumulation, which agreed with the research of Li Shuhui et al [28].

Table 9. Correlation of heavy metals in surface soil of Shouguang facility agriculture.

| Items       | As   | Hg   | Pb   | Cd   | Cr   | Cu   | Ni   | Zn   | available phosphorus | Hydrolysable nitrogen | available organic potassium matter |
|-------------|------|------|------|------|------|------|------|------|----------------------|------------------------|-----------------------------------|
| As          | 1    |      |      |      |      |      |      |      |                      |                        |                                   |
| Hg          | 0.068| 1    |      |      |      |      |      |      |                      |                        |                                   |
| Pb          | 0.107| 0.163| 1    |      |      |      |      |      |                      |                        |                                   |
| Cd          | 0.185| 0.145| 0.238| 1    |      |      |      |      |                      |                        |                                   |
| Cr          | 0.043| 0.083| 0.000| 0.307*| 1    |      |      |      |                      |                        |                                   |
| Cu          | 0.169| 0.309**| 0.1580.568"| 0.360**| 1    |      |      |      |                      |                        |                                   |
Facility agriculture featured high temperature, high humidity, high multiple cropping index, more fertilizer input, etc., with higher intensity of human disturbance than the field or other vegetable plots [28]. As investigated, fertilizers applied in Shouguang facility agriculture were mainly chicken manure, rice hull, soybean and other organic fertilizers and nitrogen, phosphorus and potassium fertilizers. Organic fertilizers were high in Cu, Zn, Cd and Hg content [36-37], compound fertilizers, especially phosphorus fertilizers, were high in Cd and Zn [36]. The above research indicated certain possible correlations between heavy metal accumulation in soil and the application of organic fertilizer, phosphate fertilizer and compound fertilizer, which also agreed basically with the research of Li Shuhui [28,38], Chen Fang [39], Zeng Xibai [40], Li Lianfang [41] and others.

4. Conclusion

(1) Heavy metals in Shouguang facility agriculture have shown a significant cumulative trend. Compared with the control and the geochemical background level of soil in Weifang, heavy metals aside from Pb and Ni exceeded the standards to different degrees; compared with HJ333-2006 and GB15618-2018, the average of heavy metal content met the standards, only Cd, Hg and Zn content in some samples exceeding the standards. According to evaluation standards of Nemerow index, 7.69%-10.77% samples of facility agriculture soil have reached the mild level of pollution, and 3.08% the moderate level.

(2) The content of heavy metals in soil of old facility agriculture (>10 years) in Shouguang is higher than that of new facility agriculture, and the heavy metal content in surface soil (0~20cm) higher than in deep soil.

(3) According to analysis of the frequency distribution of heavy metal content in surface soil, the distribution of heavy metal content is subject to exogenous heavy metals. Among them, Hg and Cd are of the highest coefficient of skewness, and most affected by exogenous sources.

(4) There are significant positive correlations among heavy metals and between heavy metals and available phosphorus, hydrolysable nitrogen, available potassium and organic matter in surface soil of facility agriculture in Shouguang.

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