

Original Research

Analysis of Soil Heavy Metal Hg Pollution Source Based on GeoDetector

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Abstract

Heavy metal mercury (Hg) is one of the most seriously polluted heavy metals in the soil of Liaocheng, China. But the source of Hg is still not clear. In this study, 85 soil samples were collected in the entire area of Liaocheng to obtain the spatial distribution of Hg. The geological accumulation index was used to evaluate the pollution in the study area. The results showed that the average value of Hg exceeded the background value of Shandong Province, and in most areas of Liaocheng, Hg was in a light-medium pollution. In addition, the paper found obvious spatial heterogeneity of Hg. So, the study area was divided into 2 parts according to the results of geological accumulation index - I_{geo} : the low-value areas and high-value areas. They were also the two layers of dependent variable in the analysis of GeoDetector. It was used to find the source of heavy metal Hg pollution based on seven influencing factors: soil pH, soil organic matter, land use type, soil subcategory, GDP, distance from roads, and distance to water. Based on the results of the GeoDetector, the main factors affecting Hg in the I_{geo} low-value area were organic matter and soil type. Thus, the source of Hg in this area was mainly natural factors such as soil parent material. While in the I_{geo} high-value area, the paper found high-density and high-polluting industries with the Point of Interest data of factories and enterprises. It indicated that waste water, waste gas, waste residue generated in the production process of these industries were the main source of Hg pollution.

Keywords: GeoDetector, soil heavy metal Hg, impact factors, pollution source

Introduction

The destructiveness and irreversibility of heavy metals to soil have become one of the most serious problems of soil pollution [1, 2]. In the past 30 years, with the rapid economic development, heavy metal pollution has become increasingly serious [3, 4]. Fuel combustion, automobile exhaust, production, and living

emissions react in heavy metals accumulated in the soil, destroy the soil texture, soil degradation [5]. Heavy metals in soil are absorbed by plants, which will be harmful to human health [6–8]. Hg is one of the most toxic pollutants. High concentration of Hg will cause ecological damage [9]. Gaseous Hg pollutants have the characteristics of moving with the atmosphere. It can be transmitted over long distances, causing large-scale pollution [10]. Therefore, it is necessary to find the source of Hg pollution in the study region. Relevant research mainly used correlation analysis,

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principal component analysis [11], and multivariate statistical analysis [12] to identify pollution sources. These methods were more effective when there was a significant linear regression between variables. Therefore, this study used geographic detectors to detect the source of Hg pollution in the study area.

The geographical detector method (GeoDetector) can identify the spatial hierarchical heterogeneity and reveal the driving force behind the research object [13]. Geographic detectors can measure the correlation between the dependent variable and the independent variable in linear and nonlinear spatial distribution. The method has been applied in many fields. Gao et al. used geographic detectors to determine the influence of selected factors on the results of remote sensing ecological index models [14]. Yang et al. used geographic detectors to reveal the influencing factors and driving mechanism of the formation of rural settlement land pattern [15]. In addition, geographic detectors are widely used in related fields such as studies of the atmosphere [16-18], soil pollution [19-21], and urban economy [22].

This paper aimed to find the source of Hg pollution in the soil in Liaocheng City. GeoDetector was used to reveal the correlation between heavy metal Hg and factors such as soil pH, soil organic matter, land use type, soil type, gross domestic product (GDP), distance from roads, and distance from water. It is important to identify the main factors affecting the sources of soil heavy metal pollution to provide a scientific basis for environmental protection and soil remediation in the study area [23-25].

Materials and Methods

Study Area

Liaocheng is in the western part of the Shandong Province, with Zhangwei River in the west, Jindi and Yellow Rivers in the southeast, and Jining, Tai'an, Jinan, Handan, and Dezhou adjacent to it. It is located at 115°16'-116°12'E and 35°47'-37°02'N. The city has a total area of 8715 square kilometers, including eight counties. In 2020, the city's GDP has reached 225.982 billion yuan. With rapid economic development, a series of environmental problems such as the deterioration of the environmental quality of scenic spots, water resource problems, and soil pollution followed [26].

Materials

The sampling points in the study area were designed in the center of 10 km × 10 km grid (Fig. 1). After sampling, the soil samples were sent to the laboratory to extract the Hg, the organic matter, soil type, soil pH. The basic data used in the study included the 2015 GDP, the 2015-2016 Shandong Provincial Statistical Yearbook

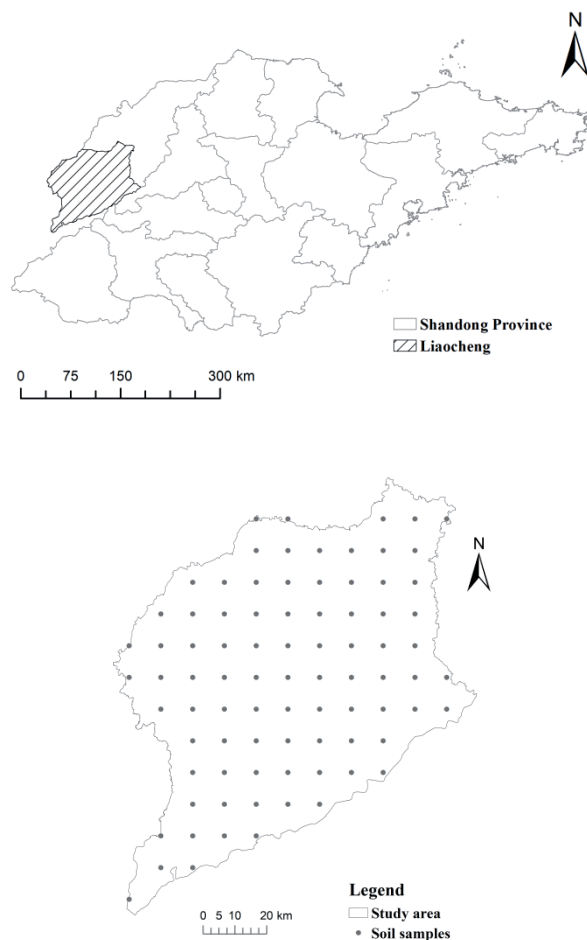


Fig. 1. Study area and location of sampling points.

(<http://www.resdc.cn/>), the third national land survey data, road data, and Point of Interest (POI) data. The POI data were obtained from the Baidu map database through python language programming. A total of 3854 location samples were imported into ArcGIS 10.3 for projection conversion. Several factors were selected for the study such as soil pH, soil organic matter, land use type, soil subcategory, GDP, distance from roads, and distance to water. GDP was calculated through the 2015 GDP data and the 2015-2016 Shandong Provincial Statistical Yearbook; the distance from road and distance to water factors were found by calculating the Euclidean distance in ArcGIS 10.3.

Methods

Geological Accumulation Index

The geological accumulation index method [27] was first proposed by the German scientist Muller to evaluate the heavy metals in sediments. This method can not only distinguish the degree of human impact on the environment, but also reflects the natural distribution of heavy metals in the soil. The calculation formula is

Table 1. Classification of pollution levels by geoaccumulation index.

I_{geo}	Level	Degree of pollution
$I_{geo} \leq 0$	0	No pollution
$0 < I_{geo} \leq 1$	1	Mild to moderate pollution
$1 < I_{geo} \leq 2$	2	Moderate pollution
$2 < I_{geo} \leq 3$	3	Moderate to heavy pollution

$$I_{geo} = \log_2[C_n / (k \times C_i)] \tag{1}$$

...where I_{geo} is the geological accumulation index of soil heavy metals; C_n is the measured value of soil heavy metals in mg/kg; C_i is the soil background value of heavy metals in the study area; and k is used to correct the irregularities in the background value of soil heavy metals that may be caused by sedimentation (generally $k = 1.5$). In addition, the potential ecological risk index is used to support the results of the geological accumulation index, as detailed in Hakanson's literature [28]. The pollution level can be divided according to the geological accumulation index (see Table 1).

Geographical Detector

The Geodetector is a model proposed by researcher Wang Jinfeng to analyze the heterogeneity of spatial stratification. The model is divided into 4 modules: factor detection, risk detection, ecological detection, and interaction detection.

Factor detection is used to detect the spatial hierarchical heterogeneity of variable Y , and the extent to which impact factor X explains the spatial differentiation of variable Y , which is measured by the q value [29, 30]. In this study, the factor detector is used to analyze the correlation between the selected impact factors and heavy metal Hg, to analyze the attribution of the pollution source. The calculation formula is

$$q = 1 - \frac{\sum_{i=1}^L N_i \sigma_i^2}{N \sigma^2} = 1 - \frac{SSW}{SST} \tag{2}$$

...where $i = 1, \dots, L$ is the division or classification of factor X or variable Y ; N_i and N are the number of units in layer i and the whole area, respectively; and are the variance of variable Y in layer i and the whole area, respectively; and $SSW =$ and $SST =$ indicate the sum of variance within the layer and the total variance of the whole area, respectively.

Results

Statistical Analysis

The statistical description of Hg is shown in Table 2. Radial basis function [31] was used to obtain the spatial

Table 2. Statistical description parameters of Hg.

Parameters	Result
Mean value (mg/kg)	0.042
Range (mg/kg)	0.008-1.635
Skewness	25.668
Kurtosis	905.95
Coefficient of Variation	1.024
Background values for Shandong Province (mg/kg)	0.019
National Level II Standards (mg/kg)	0.5

distribution of Hg (Fig. 2). The root mean square error of the model was 0.036. The average value of Hg was 0.042 mg/kg, which was 2.21 times the background value in Shandong; the maximum value was 1.635 mg/kg, which was 86 times the background value in Shandong and 3.27 times the national secondary standard, indicating that the concentration of Hg in the study area was abnormal. The area with a higher concentration than the national secondary standard had been polluted by Hg, which could damage the ecological environment. It could be seen from the skewness and kurtosis that the Hg did not conform to the normal distribution, and its coefficient of variation was much higher than 0.36 [32], indicating that the distribution of heavy metal Hg was highly variable and the soil in the study area was likely subject to human disturbance. In addition, it can be seen from Fig. 2 that the Hg was distributed

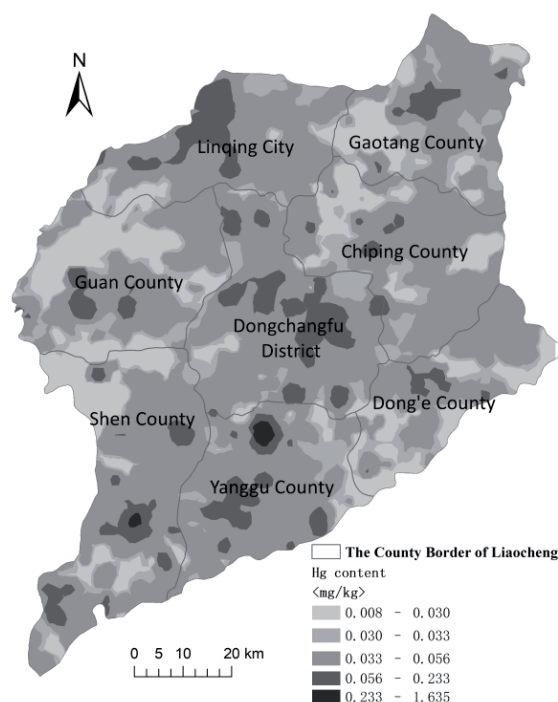


Fig. 2. Spatial distribution of heavy metal Hg.

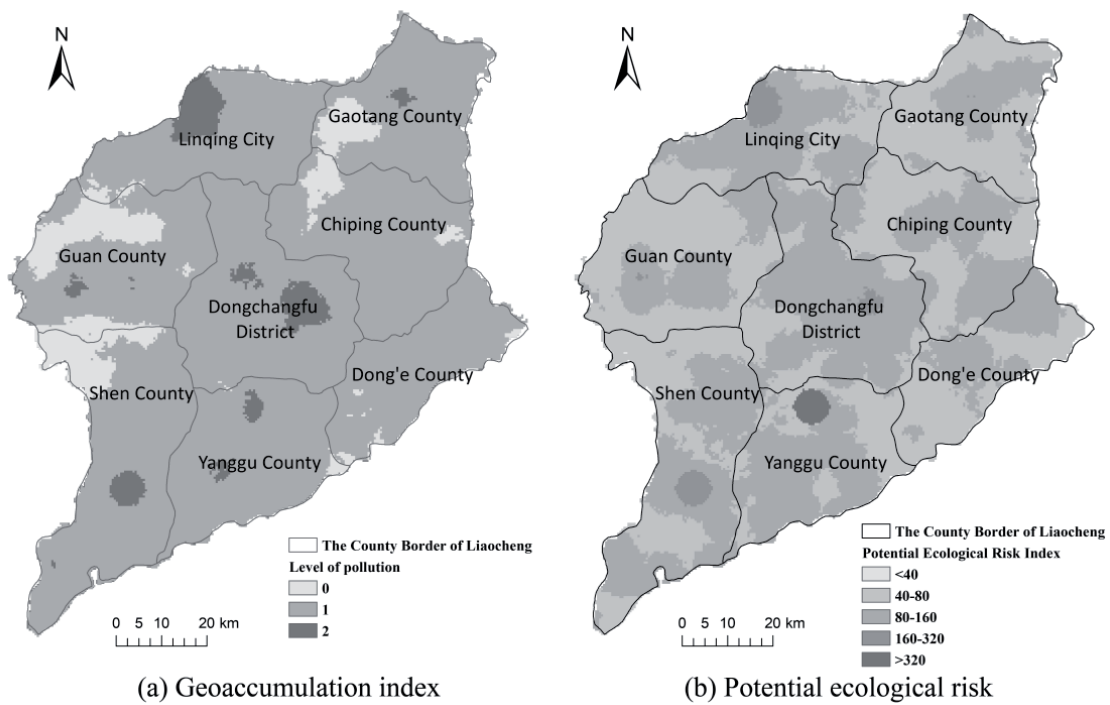


Fig. 3. Spatial distribution of heavy metal mercury pollution.

in various counties and cities throughout Liaocheng, and concentrated in Linqing, Dongchangfu District, Yanggu, and Shen County.

Analysis of Pollution Index

The Hg pollution in the study area was evaluated according to the Geoaccumulation index (Fig. 3a). In most areas of Liaocheng, Hg was found in a light-medium pollution concentration. The areas most heavily polluted by Hg were concentrated in Linqing, Dongchangfu District, and Shen County. To verify the result of the geoaccumulation index, this study used the potential ecological risk index method to evaluate the Hg pollution in the area. The potential ecological risk index of heavy metal mercury <40 is mild risk, 40-80 is medium risk, 80-160 is medium-high risk, 160-320 is high risk, ≥ 320 is highest risk. The results are shown in Fig. 3b). As shown, most areas of Liaocheng were at higher risk. The higher risks were mainly distributed in Linqing, Dongchangfu District, Yanggu County, and Shen County, which were found to have similar levels of Hg concentration as the heavily polluted areas evaluated by the geoaccumulation index.

Geodetector Analysis

Because the data used by the geodetector software (<http://geodetector.cn/>) in the calculation process were type data rather than continuous data, the dependent variable needed to be discretized. This study used the natural breakpoint method and K-means [33] as the two methods to classify the data (Table 3). The spatial distribution of each factor classification is shown in Fig. 4.

After detecting the concentration and distribution of heavy metal Hg in the soil of Liaocheng (Table 4), the q value was found to be relatively small. Based on the results of geological accumulation, the study was divided into two layers. The first layer was called the I_{geo} low-value area, or the area where the geological accumulation index level was 0 and 1. Then, the second layer was called the I_{geo} high-value area, or the area where the geological accumulation index level was 2.

Pollution Analysis of I_{geo} Low-Value Area

The research was divided into two layers and then the factor detection was performed on each layer.

Table 3. Factor classification.

Impact factors	Organic matter	Distance from road	Distance to river
Natural breakpoint	7	5	7
Impact factors	pH	GDP	
K-means	9	9	

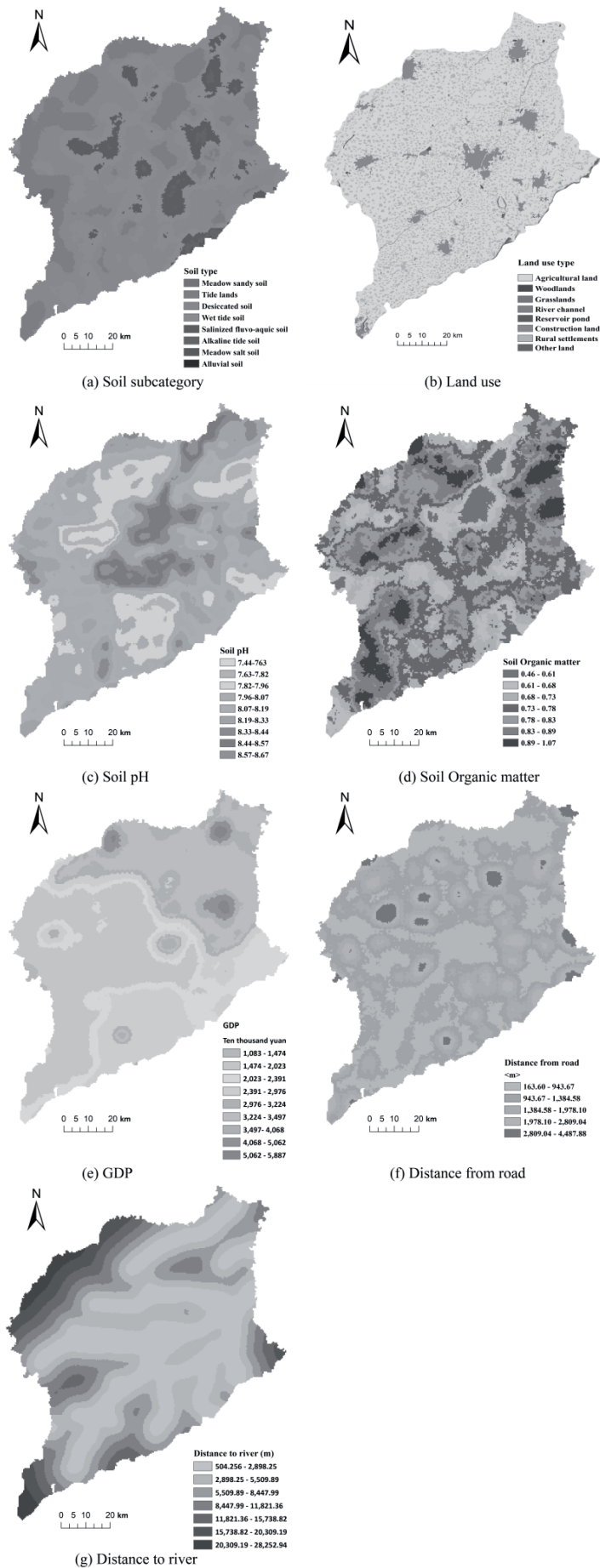


Fig. 4. Spatial distribution of various impact factors.

Table 4. Factor detection results.

Factors	Organic matter	Soil type	Soil pH	Land use type	Distance to water	Distance from road	GDP
q	0.030	0.009	0.005	0.010	0.003	0.005	0.010

Table 5. Factor detection results of I_{geo} low-value area.

Factors	Organic matter	Soil type	Soil pH	Land use type	Distance to water	Distance from road	GDP
q	0.160	0.064	0.015	0.026	0.010	0.014	0.026

The result of the I_{geo} low-value area is shown in Table 5. The impact factors were sorted according to the q value as follows: organic matter (0.160)>soil type (0.064)>GDP (0.026) = land use type (0.26)>pH (0.015)>distance from road (0.014)>distance to river (0.01). According to the results of factor detection, organic matter and soil type were the two factors most closely related to Hg in the I_{geo} low-value areas. The aforementioned two factors are natural factors, indicating that the concentration of Hg in the low-value area was mainly affected by natural factors such as soil parent material and weathering. In addition, due to the volatility of Hg and its compounds [34], Hg pollutants would diffuse through the air from the pollution source to the surrounding areas. In the I_{geo} low-value area, the main soil subcategory was fluvo-aquic soil, which has a viscous texture and high pH, making it easy to adsorb pollutants. Therefore, as shown in Fig. 3a), the moderately polluted area was surrounded by light-medium polluted areas, and only a type few non-polluted areas were found, and they were far from the moderately polluted areas.

Pollution Analysis of I_{geo} High-Value Area

For the I_{geo} high-value area, the q value of the impact factor was between 0.009 and 0.034. This might be due to the poor explanatory power of the obtained data for the distribution of heavy metal Hg in the I_{geo} high-value area. Combined with Fig. 4b) and 4e), this region had a large area of urban land, and the high GDP was concentrated in these areas. The area was economically developed and was greatly affected by human activities. Thus, the source of heavy metal Hg in the I_{geo} high-value area was analyzed directly from the location of the public enterprise.

This study used the POI data of factories and enterprises in Liaocheng and classified the POI data of various factories and enterprises using the introduction of the Liaocheng People’s Government Economic and Social Development Statistical Bulletin (<http://www.liaocheng.gov.cn/lcsq/jjsh/>). The result is shown in Fig. 5. In the I_{geo} high-value area, the density of factories and enterprises was very high. These factories and enterprises were mainly textile printing and

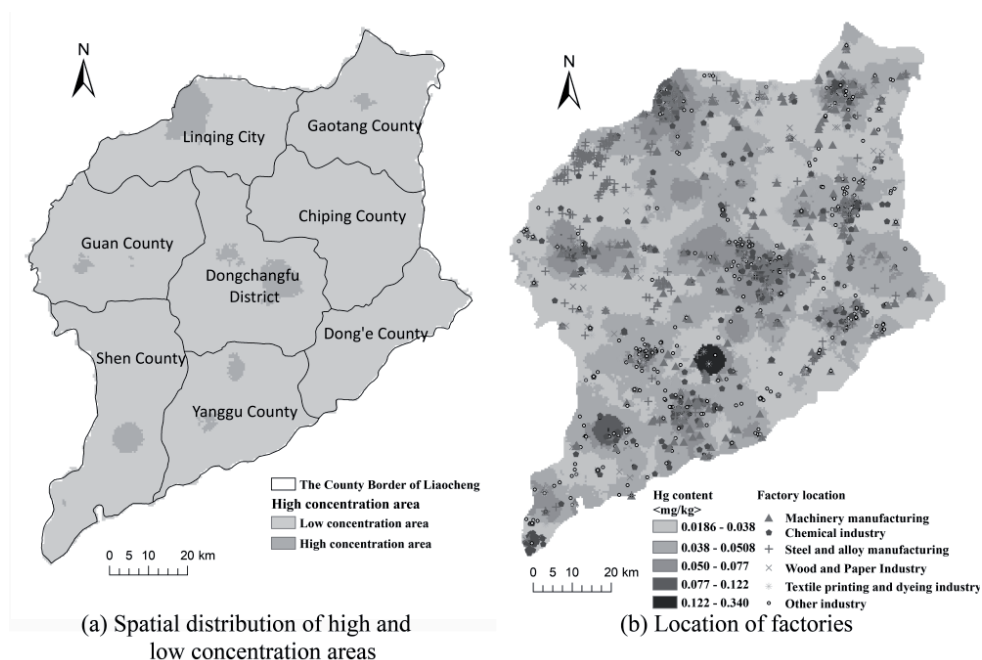


Fig. 5. Spatial distribution of industrial enterprises

dyeing, machinery manufacturing, various non-ferrous metal alloy manufacturing, chemical industry, and papermaking. These enterprises are in highly polluting industries [35]. Among them, the textile and paper industries used heavy metal Hg as the flowing cathode chlor-alkali process during production, resulting in extremely high levels of pollutant Hg emissions [36]. Machinery manufacturing, metal smelting, and chemical industry discharged the "three wastes" containing a large amount of heavy metals in the production process.

Discussion

The concentration of the heavy metal Hg in the soil of the study area ranged from 0.008 to 1.635 mg/kg, with an average value of 0.042 mg/kg, which was about twice the Shandong background value. The maximum value far exceeded the national secondary standard, indicating that the soil samples at individual sampling points exceeded the standard, causing damage to the local ecology. In addition, the coefficient of variation of the concentration of Hg was 1.024, indicating that Hg had accumulated in the study area, mainly in Linqing, Dongchangfu District, Yanggu County, and Shen County. Through the pollution index analysis, it was found that heavy metal Hg was seriously polluted in Linqing, Dongchangfu District, Yanggu County, and Shen County. These areas were the economically developed and urbanized regions in the study area, indicating that these areas were heavily influenced by humans. The study was divided into two layers according to the results of the geoaccumulation index: I_{geo} low-value area and I_{geo} high-value area. The selected seven impact factors were separated from the Hg for factor detection.

In the I_{geo} low-value areas, factor detection analysis showed that Hg was mainly affected by two natural factors, organic matter and soil type. In addition to these two natural factors, GDP and land use types also had an impact on the distribution of Hg. These two factors reflect economic development and the intensity of human activities. Hg shown in Fig. 4b) and 4e), the GDP in the I_{geo} low-value area was at a medium level, and farmland was the main land use type in this area. There was no large area of urban land in this region, but there were densely distributed rural settlements, indicating that in addition to natural factors, wastewater and waste gas generated by local residents' production, life, and agricultural activities were also sources of Hg. In addition, even though soil pH, distance from roads, and distance from rivers had a certain impact on Hg, the impact was relatively small. According to the above analysis, the heavy metal Hg found in the I_{geo} low-value area was mainly from natural factors such as soil parent material. Due to the volatile nature of Hg and the characteristics of the local economy and land use, the heavy metal Hg in these areas was affected by the proliferation of pollution sources from the I_{geo}

high-value area. At the same time, the dense rural settlements and frequent agricultural activities in this area also had an impact on the heavy metal Hg distribution.

In the I_{geo} high-value area, through POI data analysis, Hg was mainly affected by the three industrial wastes. Hg and its compounds were very volatile, allowing them to be transported over long distances through the atmosphere. Approximately 93.7% of Hg can fall back to land [37]. Hence, soil was the largest pollutant receiver of heavy metal Hg. In addition, as shown in Fig. 5b), areas with high Hg concentrations overlapped with I_{geo} high-value areas. The trends of the concentration level of Hg were basically the same as that of high-polluting industries. The above analysis showed that these high-polluting industries were the main source of heavy metal Hg pollution in the region, and these areas were mostly the central urban areas of counties and cities. Thus, it is necessary to pay attention to the pollution to prevent damage to the ecological environment of the region and the health of residents. This article had limitations in factor selection and there were few socioeconomic factors, so relevant factors should be added to continue research.

Conclusions

According to statistical analysis and pollution evaluation, Hg pollution existed in Liaocheng. In order to detect the source of Hg pollution, the study area was divided into the I_{geo} low-value area and the I_{geo} high-value area based on the results of geological accumulation. In the I_{geo} low-value area, the q values of organic matter and soil type were 0.16 and 0.064, respectively. This indicated that the main source of heavy metal Hg in the I_{geo} low-value area was soil parent material and other natural factors. However, due to the volatility of Hg and the local economic and land use conditions, the Hg in this area was also affected by the diffusion of Hg in the I_{geo} high-value area and local human activities. For the I_{geo} high-value area, the q value detected by the 7 factors was between 0.009 and 0.034. The selected factor cannot perfectly explain the source of Hg, so the analysis was carried out from the perspective of the layout of factories and enterprises. The factories and enterprises in this area are densely distributed, and most are in high-polluting industries such as textile printing and dyeing, papermaking chemicals, machinery manufacturing, and non-ferrous metal synthesis. High-polluting industries produce industrial "three wastes" with extremely high Hg content.

At present, there is no indicator that can directly and quantitatively reflect various types of enterprises to explain the source of Hg, so the analysis is limited to the consistency of spatial distribution. Hg and its compounds were volatile in this region, allowing Hg to be transmitted over long distances. Thus, the I_{geo} low-value areas surrounded high-concentration

areas, and non-polluted areas were in remote places. The heavy metal Hg content in areas with densely distributed high-polluting industries increased along with density and industry (as shown in Fig. 5b). These high-polluting industries were the main source of heavy metal Hg pollution in the area. This area is economically developed, so these pollution sources need to be treated to prevent harm to the health of residents.

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Conflict of Interest

The authors declare no conflict of interest.

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