Study on Heavy Metal Pollution Characteristics and its Health Risk Assessment from Sludge of Sewage Treatment Plants in Industrial Parks

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Abstract

Background: In order to study the characteristics of heavy metal pollution from sludge in industrial parks and the effective methods of health risk assessment, in this paper, the pollution characteristics of eight heavy metals including Zn, Cd, Pb, Hg, Cr, Ni, As, Co in sludge of sewage treatment plants in Nanjing MV Industrial Park were comprehensively evaluated and compared by using Nemerow Index Method, Muller Index Method and Matter element extension method. On this basis, the health risk assessment models of non-carcinogenic and carcinogenic heavy metals were constructed, and the health risks caused by four kinds of non-carcinogenic heavy metals Pb, Zn, Cd, Hg and four kinds of carcinogenic heavy metals Cu, As, Co, Ni contained in sludge were evaluated by using the practical data of Nanjing Industrial Park.

Results: According to the evaluation results of heavy metal pollution characteristics in sludge of sewage treatment plants in Nanjing MV Industrial Park, the order of different pollution levels under different assessment methods is as follows: the level of the Nemerow Index Method assessment was low, the level of the Muller Index Method assessment was high, and the level of Grey Clustering Method assessment was relatively moderate. In the health risk assessment of heavy metals, the results showed that the order of effects of non-carcinogenic heavy metals on health risk ranged from large to small was: Hg, Cr, Pb, and Zn, and the health risk of non-carcinogenic heavy metal in children was much higher than that in adults. Similarly, the order of effects of carcinogenic heavy metal health risk ranged from large to small was Cu, Cd, Ni and As, and the health risk of carcinogenic heavy metal in children was slightly lower than that in adults.

Conclusions: It can be seen that there is a certain degree of heavy metal pollution in the sludge of the sewage treatment plant in Nanjing MV Industrial Park, which affects the environmental quality in the park and the health risks of residents. This study verifies the assessment method of the heavy metal pollution characteristics and health risk is effective, and its research results have a guiding role in the management of heavy metal pollution in the sludge of the sewage treatment plant in Nanjing MV Industrial Park. It also has significant theoretical support for the government to formulate the eco-environmental quality standards and related policies of industrial parks.

Keywords: Sludge; Heavy metal pollution; Pollution characteristics assessment; Health risk assessment; Industrial parks; Sewage treatment plants

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1. Background

With the rapid development of China's industrial parks, the sewage treatment plants in industrial parks have been building in order to deal with industrial production and living sewage discharge according to the requirements of ecological industrial park construction. The use of the sewage treatment plants purifies the water and reduces the direct discharge of sewage from the industrial park into the Yangtze River, thus protecting the ecological environment to a large extent (Zhang et al., 2013). However, along with the operation of the industrial park sewage treatment plants, while continuously purifying the water quality, it is also constantly producing a large amount of sludge (Li et al., 2019). Sludge produced by these sewage treatment plants is a biologically active mixture containing a large number of bacteria, germs and toxic substances (Yang et al., 2019). According to the statistics of China's urban and rural construction statistical yearbook, by the end of 2018, China had 2,300 urban sewage treatment plants, and its sewage treatment capacity had increased from 38 billion M$^3$ in 2010 to 59.1 billion M$^3$; its annual sewage treatment capacity had increased from 31.2 billion M$^3$ in 2010 to 47.9 billion M$^3$, and China's urban sewage treatment rate rose from 82.32 percent to 95.42 percent. With the increase of urban and rural sewage discharge scale and treatment capacity in China, about 10-30 tons of sludge can be produced in every 10,000 M$^3$ sewage. In 2010, the minimum size of sludge produced by sewage treatment plants in China's urbanization was 31.2 million tons, and the maximum size was 93.6 million tons, with an average value of about 62.4 million tons. By the end of 2018, the minimum size of sludge increased to 47.9 million tons, and the maximum size increased to 143.7 million tons, the average was about 95.8 million tons. Based on average, sludge from urban and rural sewage treatment plants increased by 53.53 percent during the eight-year period, with an average annual growth rate of 6.69 percent.

Nanjing MV Industrial Park is located in the high-tech development zone of Pukou District, Nanjing, China. The sewage treatment plants concentrate on the treatment of wastewater from the production and living of
enterprises in the industrial park, with an average daily capacity of 22,000 M³ and an annual production of about 16,500 tons of sludge. There are many kinds of heavy metal in sludge, including zinc (Zn), lead (Pb), copper (Cu), cadmium (Cd), chromium (Cr), nickel (Ni), mercury (Hg), and arsenic (As). The heavy metal toxicity, such as cadmium (Cd), chromium (Cr), arsenic (As), nickel (Ni), can cause cancer in people who inhale too much these substances, and excessive intake of other non-carcinogenic heavy metals may also pose health risk (Yang et al., 2019). Sludge produced by sewage treatment plants of Nanjing MV Industrial Park is mostly used as fertilizer by farmers around the industrial park, a few sludge is landfill treatment, and individual heavy metal high pollution sludge is incinerated. No matter which treatment method is adopted, the bacteria, germs and heavy metals in the sludge will pollute the environment to a certain extent, threatening the health and even life of the people concerned in the industrial park (Yang et al., 2019). Heavy metal pollution has the characteristics of long time and great harm, therefore, it is very important and urgent to study the characteristics of heavy metal pollution and health risk assessment in sewage treatment plants in industrial parks.

The study of heavy metal pollution began in western developed countries. In the early 20th century, the scientific committee of the British Medical Association explained the general toxic effects of heavy metals after subcutaneous injection and the poisoning of paint volatiles and began to set standard (Moore et al., 1913). During this period, doctors in the United States found that excessive intake of zinc (Zn) into the human body will cause permanent damage to human tissues and excessive intake of nickel (Ni) into the human body will cause damage to nerve endings muscle fibers (Salant and Mitchell, 1915). French doctors treating patients with syphilis have found that excessive intramuscular injection of heavy metal salts can cause local medical accidents, and that excessive intake of heavy metal salts into the body can damage muscle tissue (Gammel, 1928). The study on the risk assessment of heavy metal pollution in developed countries began in the late 1940s. In the treatment of early syphilis, the United States hospitals tried to improve the treatment effect by combining heavy metals. However, treatment accidents occurred in the course of using heavy metal...
combination therapy, and people began to consider strengthening the evaluation of the effect of heavy metal combination therapy, which is the first overseas study on the assessment of the impact of heavy metals on human health (Heller, 1946). Since then, for the treatment of early syphilis in hospitals in developed countries, the combination of heavy metals is often used, such as penicillin combined with heavy metal arsenic (As) treatment of early syphilis evaluation (Goldmann et al., 1947), asfenamin combined with heavy metals treatment of early syphilis evaluation (Thompson and Smith, 1950) and so on. Study on heavy metal pollution in sludge and its health risk assessment began in the early 1960s. Jenkins and Cooper (1964) analyzed the present situation of heavy metals in sludge, they found that heavy metal intake in sludge had a negative impact on human health, reminded people to pay attention to heavy metal pollution in sludge. Brown et al. (1973) studied the removal of heavy metals and their removal efficiency in six municipal sewage treatment plants in the United States, and focused on the analysis of heavy metal removal effect in Kansas City sewage treatment plants. After decades of development, the research on heavy metal pollution and its health risk assessment in developed countries has gradually moved towards standardization, and the research focus has gradually shifted to heavy metal pollution in industrial production (Ukah et al., 2019; Udayanga et al., 2019), especially in industrial parks (Selvam et al., 2017; Pobi et al., 2019). It can be seen that although the western developed countries have relatively deep research on heavy metal pollution and its health risk assessment, there are relatively few studies on heavy metal pollution in industrial parks, especially on heavy metal pollution and health risk assessment in sludge of sewage treatment plants in industrial parks.

China's research on heavy metal pollution and its health risk assessment started relatively late, and the earliest research began in the early 21st century. Chang et al. (2000) analyzed the relationship between heavy metal pollution and human health, and clearly pointed out that excessive intake of heavy metals by human body is easy to produce serious health problems and even pose a threat on life. Subsequently, domestic research on heavy metal pollution and its health risk assessment has been gradually carried out, and its
contents and scope have been gradually expanded. The related research mainly covers the following areas: heavy metals contained in vegetable soil, which can pose a threat to human health through the production, transportation and consumption of vegetables (Ding and Pan, 2003), heavy metal pollution in water bodies and its health risk assessment (Gao et al., 2004), heavy metal pollution in acid rain and its health risk assessment (Xu et al., 2008), heavy metal pollution and health risk assessment in mining areas (Huang et al., 2009), heavy metal pollution in water sediments and its health risk assessment (Deng et al., 2011), heavy metal pollution and its health risk assessment in municipal solid waste incineration (Zhang et al., 2013), and heavy metal pollution in sludge from municipal sewage treatment plants and its health risk assessment (Wei et al., 2012). Since 2010, domestic research on heavy metal pollution and its health risk assessment has gradually shifted to industrial parks, focusing on: dust and heavy metal pollution around industrial parks and their risk assessment (Ren et al., 2012), heavy metal pollution in industrial park soils and its health risk assessment (Peng et al., 2013), heavy metal pollution and its health risk assessment in the atmosphere of industrial parks (Liang et al., 2014), heavy metal pollution and its health risk assessment in wastewater discharge of industrial parks (Yao et al., 2016), and heavy metal pollution and health risk assessment in sludge of sewage treatment plants in industrial parks (Zhou et al., 2018). China's current research on heavy metal pollution and its health risk assessment of sewage treatment plants in industrial parks shows an upward trend, and the research focus has also begun to turn to the evaluation method and its application research (Chang et al., 2019; Mao et al., 2020).

From the above literature review, it can be clearly seen that the academic research results on heavy metal pollution and its health risk assessment are relatively large, and this research is mainly to explore the application in new situations and some special fields. However, domestic research on heavy metal pollution and its health risk assessment in industrial parks is relatively few, while the research on heavy metal pollution and its health risk assessment in sewage treatment plant sludge is even less, which is not suitable for the development of industrial parks in China. China’s industrial parks have become the main force in China’s
economic development, and the contribution rate of industrial parks in China’s economic development has been close to 40%. The rapid development of China’s industrial parks has gradually exposed some defects, mainly manifested in: the lag of environmental pollution control, the relative shortage of investment in environmental pollution control and the limitation of environmental pollution control ability. Sewage treatment plants are generally built in China’s industrial parks, however, the sewage treatment effect is not very ideal due to many reasons. The phenomenon of heavy metal content exceeding the standard in sludge is more common, and sludge accidents occur frequently. Therefore, in this case, it is of great theoretical significance and practical value to study the heavy metal pollution characteristics and health risk assessment in sludge of sewage treatment plants in industrial parks.

2. Materials and Methods

2.1 Sludge Sample Collection and Analysis

The industrial enterprises in Nanjing MV industrial park use a common sewage treatment plant with an annual sewage treatment capacity of 8 million M³ and a production of 16,500 tons of sludge annually. In order to evaluate the pollution characteristics and health risk of heavy metals in sludge produced by the sewage treatment plant, it is necessary to sample the sludge produced by sewage treatment plant to determine the content of heavy metals in sludge and the degree of health risk. The principles of full coverage, combination of randomization and judgment and collection of valid samples are adopted to make the samples representative.

In the dewatering plant of the selected sewage treatment sludge, the standard sample number is selected at the end of the dewatering machine of final sludge production, and at the sludge piling site, the standard sample number is selected at regular intervals according to the characteristics of sludge piling up. The standard sample selected in this paper is a mixed sample, which contains all heavy metals to be measured. The mass of the mixed sample is 500 grams; the sampling period was between 1 and 31 December 2019. One sludge sample is collected every day for five days before and after this month, and one sludge determination sample is collected
every other day from 6 to 26 days. A total of 20 sludge determination samples are collected. The sludge was sampled and wrapped with foil paper, each sample was numbered according to the order of sampling from 001 to 020, and finally the sample was sent to the laboratory according to the procedure. The technical personnel of the laboratory carry on the necessary technical treatment to the sample according to the heavy metal measurement request, mainly carrying on the drying treatment, the grinding, the sieve and the digestion to the determination sample. The qualified samples will be carried on the standardized determination for heavy metal content by using professional measuring equipment. The results of measurement are detailed in Table 1.

| No.  | Pb   | Zn   | Cu   | Hg   | Cr   | As   | Cd   | Ni   |
|------|------|------|------|------|------|------|------|------|
| 001  | 38.48| 119.48| 50.34| 0.18 | 68.28| 16.32| 0.31 | 39.37|
| 002  | 39.49| 125.38| 52.39| 0.15 | 67.32| 15.28| 0.28 | 38.31|
| 003  | 36.27| 121.27| 55.37| 0.11 | 65.39| 17.62| 0.21 | 41.28|
| 004  | 37.29| 130.25| 50.28| 0.13 | 60.26| 18.29| 0.22 | 37.49|
| 005  | 33.49| 128.32| 48.23| 0.16 | 58.38| 15.84| 0.18 | 35.28|
| 006  | 31.29| 101.28| 45.98| 0.15 | 56.39| 13.29| 0.19 | 36.72|
| 007  | 29.37| 93.29 | 49.32| 0.14 | 55.38| 14.39| 0.2  | 39.31|
| 008  | 35.48| 96.28 | 51.29| 0.17 | 58.28| 15.39| 0.22 | 40.28|
| 009  | 34.38| 65.39 | 53.28| 0.18 | 59.62| 16.39| 0.25 | 44.74|
| 010  | 35.18| 108.29| 59.64| 0.2  | 58.39| 18.29| 0.21 | 40.82|
| 011  | 33.85| 113.29| 61.28| 0.21 | 61.28| 21.38| 0.16 | 38.37|
| 012  | 32.39| 115.38| 56.32| 0.23 | 62.38| 20.37| 0.15 | 35.39|
| 013  | 33.49| 102.53| 55.26| 0.21 | 60.29| 18.39| 0.17 | 30.38|
| 014  | 36.39| 99.25 | 53.29| 0.19 | 63.26| 17.39| 0.21 | 33.48|
| 015  | 35.98| 115.18| 56.92| 0.17 | 61.39| 16.39| 0.22 | 38.94|
| 016  | 37.62| 1.28  | 51.29| 0.22 | 70.36| 19.28| 0.26 | 40.21|
| 017  | 38.39| 98.29 | 54.63| 0.23 | 66.35| 18.39| 0.25 | 41.38|
| 018  | 31.29| 97.29 | 52.39| 0.18 | 59.38| 17.28| 0.19 | 40.89|
| 019  | 29.96| 89.28 | 55.59| 0.19 | 57.62| 20.21| 0.21 | 39.78|
| 020  | 34.49| 92.39 | 54.26| 0.2  | 55.63| 15.29| 0.22 | 40.32|
| Average| 34.73| 100.67| 50.64| 0.18 | 58.37| 16.41| 0.22 | 38.64|
| Standard Deviation| 2.8758| 28.1822| 3.8235| 0.0331| 4.3740| 2.1597| 0.0397| 3.1933|
| Coefficient of Variation| 0.0828| 0.2799| 0.0755| 0.1838| 0.0749| 0.1316| 0.1803| 0.0826|

2.2 Assessment Model Construction

2.2.1 Heavy Metal Pollution Assessment Model

The sludge of sewage treatment plants in industrial parks contains a lot of heavy metals, and the
pollution of heavy metals is different from that of organic compounds. Organic compounds pollution can be reduced or defused by physical, chemical or biological purification in nature's ecosystem, depending on the self-recovery function of the ecosystem. While heavy metal pollution has the characteristic of enrichment, and the ecosystem can not do anything about heavy metal pollution. Therefore, heavy metal pollution is more threatening to the natural environment and human health and life. The study of heavy metal pollution assessment in western developed countries began in the late 1960s. German scientists Muller first proposed and used quantitative indicators Geoaaccumulation index, studying the extent of heavy metal pollution in sediments and other substances, which were later called "Muller index"(Muller,1969). In the early 1970s, American environmentalists Nemerow NL first used the comprehensive pollution index method to assess the status of river pollution in the United States in his book Scientific Analysis of River pollution, which was later called "Nemerow index"(Nemerow, 1974). Swedish scientists Hakanson combined with the ecological effects of heavy metal elements, environmental effects and toxicological theory to assess the ecological hazards caused by heavy metal pollution (Hakanson, 1980). By the 1990s, western countries had gradually formed a traditional assessment method system for heavy metal pollution assessment. Subsequently, some scholars in China, based on the assessment method of heavy metal pollution in the west, introduced entropy weight method, coefficient of variation method, fuzzy mathematics and other methods to comprehensively assess the pollution status of heavy metals (Li et al., 2012; Ma et al., 2017).On the basis of previous studies, this paper selects Muller Index Method and Nemerow IndexMethod and introduces Grey Clustering Method to comprehensively evaluate the characteristics of heavy metal pollution in sludge of the sewage treatment plant in Nanjing MV Industrial Park.

(1) Nemerow Index Method. The model takes the form of a single factor index and a composite index. Single factor index model refers to the method of determining the main heavy metal pollutants and their harm degree by using the index of single factor. It is a pollution index expressed by the ratio between the measured
values of heavy metal content and the evaluation criteria, which reflects the pollution degree of heavy metals. Assume that \( R_i \) represents the single factor pollution index of the \( i \)th heavy metals in sludge, \( C_i \) represents the laboratory measurements of the \( i \)th heavy metal content in sludge, \( S_i \) represents the evaluation standard value for the content of the \( i \)th heavy metal in sludge, so the single factor pollution index of heavy metals in sludge of sewage treatment plants in industrial parks can be expressed as:

\[
R_i = C_i \cdot S_i^{-1}
\]  

(1)

The above single factor pollution index model can only reflect the pollution degree of a specific heavy metal, and can not fully reflect the comprehensive pollution degree of all heavy metals in sludge of sewage treatment plants. Therefore, it is necessary to construct the heavy metal comprehensive pollution index. In Nemerow Index Method, the single factor index mean value and the highest value are taken into account to highlight the heavy metal pollution. Assume that \( R_c \) represents the total pollution index of heavy metals in sludge, \( \bar{R} \) represents the average value of single factor pollution index and \( \max (R_i) \) represents the maximum value of single factor pollution index, then:

\[
R_c = \left[ \frac{1}{2} \left( \left( \sum_{i=1}^{n} \left( \frac{C_i}{S_i} \right) / n \right)^2 + \max \left( \frac{C_i}{S_i} \right)^2 \right) \right]^{1/2}
\]  

(2)

The degree of heavy metal pollution in sludge of sewage treatment plants in industrial parks can be sorted by using the above formula (1), and the overall comprehensive pollution condition of sludge can be judged by using formula (2). The importance of heavy metal pollution in sludge is regarded equally in Nemerow Index Method, the comprehensive pollution index is expressed by the root of the sum of square and maximum index square of single pollution index, and the relative weight of single index is not considered. If the relative weight of a single index is represented by \( \xi_j \) (The calculation method will be detailed in Grey Clustering Method), the comprehensive pollution index of heavy metals can be revised to:

\[
R_c = \left[ \frac{1}{2} \left( \left( \sum_{i=1}^{n} \left( \xi_j \cdot \frac{C_i}{S_i} \right) / n \right)^2 + \max \left( \xi_j \cdot \frac{C_i}{S_i} \right)^2 \right) \right]^{1/2}
\]  

(3)
According to the Nemerow quality classification standard, the assessment criteria for the pollution degree of heavy metal single factor pollution index and comprehensive pollution index in sludge of sewage treatment plants in industrial parks are shown in Table 2.

| Serial Number | Pollution Level | Single Factor Pollution Index | Comprehensive Pollution Index | Characteristics |
|---------------|-----------------|--------------------------------|-------------------------------|-----------------|
| 1             | Level I         | $R_i \leq 0.70$               | $R_i \leq 0.70$               | No pollution    |
| 2             | Level II        | $0.7 \leq < 1$ $R_i$         | $0.7 \leq < 1$ $R_i$         | Micro-pollution |
| 3             | Level III       | $1 \leq < 2$ $R_i$           | $1 \leq < 2$ $R_i$           | Mild contamination |
| 4             | Level IV        | $2 \leq < 3$ $R_i$           | $2 \leq < 3$ $R_i$           | Moderate pollution |
| 5             | Level V         | $R_i > 3$                     | $R_i > 3$                     | Heavy pollution |

(2) Muller Index Method, also known as Muller Geoaccumulation index method, is a method for assessing the extent of heavy metal contamination in sludge from an environmental geochemical perspective. In this method, human pollution, environmental geochemical background value and industrial background are considered. Therefore, it has been widely used in the assessment of heavy metal pollution degree in sludge.

Assume that $lgeo$ represents Muller Geoaccumulation index, $C_i$ represents the measured value of the $i$th heavy metal content in sludge, $B_i$ represents the pre-industrial geochemical background of $i$th heavy metals in sludge, $k$ represents the influence coefficient of the pre-industrial geochemical background value, and Muller Index model can be expressed as follows:

$$lg eo = \log_2 \left[ C_i \cdot (kB_i)^{-1} \right]$$  \hspace{1cm} (4)

$B_i$ requires for non-polluting values, and no industrial and no anthropogenic environmental impact is non-valuable. Therefore, industrialization and the impact of human behavior on the environment are considered in $B_i$. The influence coefficient of the background value of pre-industrial geochemistry is 1.5 in this paper according to Wen et al (2016) and Li et al (2019). The assessment standard for the degree of Muller Geoaccumulation index pollution according to the Muller quality classification standard are shown in Table 3.
Table 3 Muller Index Classification Standard of Heavy Metal Pollution in Sludge of Sewage Treatment Plants in Industrial Parks

| Serial number | Cumulative index (R_M) | Pollution level | Pollution level status       |
|---------------|------------------------|-----------------|------------------------------|
| 0             | \( lg e_0 \leq 0 \)   | Level 0         | No pollution                 |
| 1             | \( 0 \leq lg e_0 < 1 \) | Level I         | Mild contamination           |
| 2             | \( 1 \leq lg e_0 < 2 \) | Level II        | Moderate pollution           |
| 3             | \( 2 \leq lg e_0 < 3 \) | Level III       | Medium-strength pollution    |
| 4             | \( 3 \leq lg e_0 < 4 \) | Level IV        | Intensity pollution          |
| 5             | \( 4 \leq lg e_0 < 5 \) | Level V         | Strong-polarity pollution    |
| 6             | \( lg e_0 \geq 5 \)    | Level VI        | Extreme intensity pollution  |

(3) Grey Clustering Method. This method was developed by Professor Deng Julong, a famous Chinese mathematician, in the 1980s on the basis of the "Grey Box" theory. According to the gray correlation matrix or the whitening weight function of gray number, a class of observation indexes or objects can be clustered into several defined categories. The gray whitening weight function clustering is used to detect whether the observed objects belong to different categories set in advance(Deng,1982). This paper introduced it into the evaluation of heavy metal pollution characteristics in sludge of sewage treatment plants in industrial parks, hoping to have better health risk evaluation effect of heavy metal pollution. In this paper, gray whitening weight function clustering is used to check whether the observed objects belong to different categories set in advance. There are \( n \) clustering objects, \( m \) clustering indexes, \( s \) different gray classes. According to the index sample value \( X_{ij}(i=1,2,\ldots,n ; j=1,2,\ldots,m) \), the \( i \)th object is grouped into the \( k \) gray category (\( k=1,2,\ldots,s \)), which is called grey clustering. The specific method is to use the whitening weight function \( e_i^k(X_{ij}) \) to calculate the membership degree of the evaluation index. \( K \) represents the heavy metal pollution grade category in sludge of the sewage treatment plants in the industrial parks, and five levels are set in the evaluation of the characteristics of heavy metal pollution. Using the membership function and the relative weight value of the evaluation index, the index variable weight clustering coefficient of the relative gray evaluation grade \( k \) of the evaluation object \( i \) is calculated, and the evaluation grade is determined by taking the big principle. For the five levels of heavy metal pollution characteristic evaluation of sludge, the following membership functions are constructed.
(i) When \( k = 1 \), its membership function is expressed as follows:

\[
e_j^i(X_y) = \begin{cases} 
1 & X_y \leq S_{ik} \\
\left( S_{ik} - X_y \right) \left( S_{ik} - S_{ik} \right)^{-1} & S_{ik} < X_y < S_{ik} \\
0 & X_y \geq S_{ik} \end{cases}
\]  \hspace{1cm} (6)

(ii) When \( k = 2, 3, 4 \), its membership function is expressed as follows:

\[
e_j^i(X_y) = \begin{cases} 
0 & X_y \leq S_{ik} \leq X_y \leq S_{ik} \\
1 & S_{ik} < X_y < S_{ik} \\
\left( S_{ik} - X_y \right) \left( S_{ik} - S_{ik} \right)^{-1} & S_{ik} < X_y < S_{ik} \end{cases}
\]  \hspace{1cm} (7)

(iii) When \( k = 5 \), its membership function is expressed as follows:

\[
e_j^i(X_y) = \begin{cases} 
1 & X_y \geq S_{ik} \\
\left( X_y - S_{ik} \right) \left( S_{ik} - S_{ik} \right)^{-1} & S_{ik} < X_y < S_{ik} \\
0 & X_y \leq S_{ik} \end{cases}
\]  \hspace{1cm} (8)

In the formula, \( e_j^i(X_y) \) represents the degree of membership of the \( j \)th pollution index of the \( i \)th sampling point to the environmental pollution status of the \( k \)th grade, also known as the whitening weight function. \( X_y \) represents the actual measured value of the \( j \)th heavy metal index of the \( i \)th sampling point, and \( X_{ik} \) represents the evaluation standard value of the \( k \)th pollution category of the \( i \)th sampling point. In order to calculate the grey clustering evaluation results, we must determine the weight value of the evaluation index. The relative weight value is determined by expert investigation, hierarchy analysis, and entropy weight and so on. In this paper, the entropy weight method is used to determine the relative weight value of the evaluation index. The entropy theory was first founded by German physicist Rudolf Julius Emanuel Clausius in 1856. In 1948, the famous communication scientist Shannon CE and mathematician Norbert Wiener introduced entropy theory into information science, and put forward the concept of information entropy. Entropy is an objective weighting method, which can reduce the influence of subjective opinions on the weight determination of evaluation indicators. The raw data matrix of the evaluation indicators can be expressed as:
The heavy metal pollution characteristic evaluation index in sludge of sewage treatment plants in industrial parks is a reverse index, so it is better to select the index with low evaluation value. \( y_{ij} \) represents the standardized value, and the standardized formula can be expressed as:

\[
y_{ij} = \left( \max \left( x_{ij} \right) \right) \cdot \left( \max \left( x_{ij} \right) - \min \left( x_{ij} \right) \right)^{-1}
\]

\( Y \) represents the standardized matrix of heavy metal pollution characteristics evaluation index in sludge of sewage treatment plants in industrial parks. According to the standardization calculation formula, the standardized matrix can be expressed as:

\[
Y = \begin{bmatrix}
  y_{11} & y_{12} & \cdots & y_{1n} \\
  y_{21} & y_{22} & \cdots & y_{2n} \\
  \vdots & \vdots & \ddots & \vdots \\
  y_{m1} & y_{m2} & \cdots & y_{mn}
\end{bmatrix}
\]

\( H_i \) represents the entropy of the evaluation index. According to the definition of entropy, when the number of evaluation index is \( m \) and the number of evaluation object is \( n \), the value of entropy can be expressed as:

\[
H_i = \left( -\sum_{j=1}^{m} e_{ij} \ln e_{ij} \right) \cdot \left( \ln m \right)^{-1}
\]

In above formula, \( e_{ij} = y_{ij} \cdot \left( \sum_{j=1}^{m} y_{ij} \right)^{-1} \), so \( \ln e_{ij} \) is meaningless when \( e_{ij} = 0 \). After translation treatment of \( e_{ij} \), the formula can be expressed as:

\[
e_{ij} = 1 + y_{ij} \cdot \left( \sum_{j=1}^{m} \left( 1 + y_{ij} \right) \right)^{-1}
\]

\( \xi_{ij} \) represents the relative weight of the heavy metal pollution characteristics evaluation index in sludge of sewage treatment plants in industrial parks. The relative weight of the evaluation index can be easily determined according to the definition of entropy and entropy weight. The calculation formula is as follows:

\[
\xi_{ij} = \left( 1 - H_i \right) \cdot \left( n - \sum_{i=1}^{n} H_i \right)^{-1}
\]
evaluation grade $k$. According to the grey cluster evaluation theory, the grey cluster evaluation result model can be expressed as:

$$
\sigma^k_i = \sum_{j=1}^{m} e^j \left( X^j \right) \cdot \xi_j
$$

(15)

According to the principle of maximum membership degree, the maximum value of the clustering coefficient lies in the grey $k$th class, which is the class of the clustering objects:

$$
\sigma^k_i = \max_{1 \leq k \leq 5} \{ \sigma^k_i \}
$$

(16)

In order to evaluate the pollution characteristics of heavy metals in sludge of sewage treatment plants in industrial parks by Grey Clustering model, the evaluation standard of heavy metal pollution level should be determined first. On the basis of the research results of domestic environmental management experts (Zhang et al., 2017; Zhang et al., 2018), referring to China's Urban Sewage Treatment Sludge (GB T24188-2009), Request for Comments on Technical Standard for Sludge Treatment of Urban Sewage Treatment and Urban Sewage Treatment Pollutant Discharge Standard (GB18918-2016), the evaluation standard of heavy metal pollution characteristics in sewage treatment plant of Nanjing MV Industrial Park is detailed in Table 4.

| Table 4 Evaluation Standard of Heavy Metal Pollution in Sludge of Sewage Treatment Plants in Industrial Parks |
|---|---|---|---|---|---|---|---|---|
| k | Pollution Level | Pb | Zn | Cu | Hg | Cr | As | Cd | Ni |
| 1 | Level I | 0-25 | 0-75 | 0-30 | 0-0.15 | 0-60 | 0-10 | 0-0.20 | 0-30 |
| 2 | Level II | 25-150 | 75-150 | 30-120 | 0.15-0.30 | 60-90 | 10-15 | 0.20-0.30 | 30-50 |
| 3 | Level III | 150-350 | 150-350 | 120-280 | 0.30-0.5 | 90-210 | 15-30 | 0.30-0.60 | 50-100 |
| 4 | Level IV | 350-500 | 350-500 | 280-400 | 0.5-1.0 | 210-300 | 30-40 | 0.60-1.00 | 100-200 |
| 5 | Level V | >500 | >500 | >400 | >1.00 | >300 | >40 | >1.00 | >200 |

2.2.2 Health Risk Assessment Model Construction of Heavy Metal Pollution

The health risk assessment of heavy metal pollution in sludge of sewage treatment plants in industrial parks should be based on the factors including heavy metal content in sludge, the dosage of heavy metal ingested by relevant personnel in different ways and the standard of heavy metal endangering human health. Heavy metals in sludge of the sewage treatment plant in industrial park can threaten the health of the relevant
population by breathing inhalation, hand intake and skin contact. According to the dosage and properties of heavy metals contained in sludge, the four heavy metals $P_{b}$, $Z_{n}$, $C_{a}$, $H_{g}$ is not carcinogenic although harmful to human health, which are called non-carcinogenic heavy metals. Excessive intake of the four heavy metals $C_{r}$, $A_{i}$, $C_{d}$, $N_{i}$ contained in sludge can cause cancer, which are called carcinogenic heavy metals. According to the latest results of health risk assessment of heavy metal pollution at home and abroad, combined with the present situation of heavy metal pollution in sludge of sewage treatment plant in Nanjing MV Industrial Park, this paper analyzes the construction of health risk assessment model of non-carcinogenic heavy metal and carcinogenic heavy metal in sludge of sewage treatment plants in industrial parks respectively (He et al., 2016; Li et al., 2019).

(1) Construction of non-carcinogenic heavy metal health risk assessment model. There are four non-carcinogenic heavy metals $P_{b}$, $Z_{n}$, $C_{a}$, $H_{g}$ in the sludge of the sewage treatment plants in industrial parks. The health risk of such heavy metals to the contact is assessed by using the human exposure assessment model proposed by the EPA of the United States (Friberg, 1982). The main ways to intake heavy metals in human body are respiratory inhalation (BI), hand and mouth intake (HMI) and skin contact (SC). $DED_{ij}$ represents the average daily exposure dose ($mg / m^3 . d$) of the $j$th intake method of $i$th non-carcinogenic heavy metals. The measured value of heavy metal content (mg / kg) is denoted by $C$. The inhalation rate ($m^3 / d$) of non-carcinogenic heavy metal polluters is denoted by $NRR$. The annual exposure amount ($d / a$) is denoted by $AE$. The exposure period ($a$) is denoted by $YE$. Heavy metal particulate emission factor ($m^3 / kg$) is denoted by $PEF$. The Average body weight (kg) is denoted by $AW$. Average exposure time (d) is denoted by $NAT$. The intake rate (mg / d) is denoted by $IR$. The exposed skin area ($cm^2$) is expressed by $ESA$. The skin adhesion (mg / $cm^2$.d) is expressed by $SA$, and the skin absorption factor is expressed by $SAF$. The daily average exposure dose assessment model of heavy metals in sludge of sewage treatment plants in industrial parks can be expressed as:
\[
\begin{align*}
DE_{\text{in}} &= C \cdot NRR \cdot AE \cdot YE \cdot \left(PEF \cdot AW \cdot NAT\right)^{-1} \\
DE_{\text{inh}} &= C \cdot IR \cdot AE \cdot YE \cdot \left(AW \cdot NAT\right)^{-1} \cdot E - 6 \\
DE_{\text{isk}} &= C \cdot ESA \cdot SA \cdot SAF \cdot AE \cdot YE \cdot \left(AW \cdot NAT\right)^{-1} \cdot E - 6
\end{align*}
\]

\(NCED_{\text{j}}\) represents the non-carcinogenic health risk index for the \(i\)th heavy metal under the \(j\)th exposure pathway. \(RfD_{\text{j}}\) represents the corresponding reference dose for the \(i\)th heavy metal under the \(j\)th exposure pathway. \(NCRQ_{\text{ij}} = \left(NCED_{\text{ij}} \right) \cdot \left(RfD_{\text{ij}} \right)^{-1}\) represents the non-carcinogenic risk quotient for the \(i\)th heavy metal under the \(j\)th exposure pathway, and \(NHRI_{\text{ij}}\) represents the non-carcinogenic health risk index for the \(i\)th heavy metal under the \(j\)th exposure pathway, then:

\[
NHRI_{\text{ij}} = \sum_{j=1}^{m} \sum_{i=1}^{n} NCRQ_{\text{ij}} = \sum_{j=1}^{m} \sum_{i=1}^{n} \left(NCED_{\text{ij}} \right) \cdot \left(RfD_{\text{ij}} \right)^{-1}
\]

The above formula is the health risk assessment model of non-carcinogenic heavy metals in sludge of the sewage treatment plants in industrial parks. When \(NHRI \leq 1\), the heavy metals in the sludge do not pose a health risk to the exposed population. When \(NHRI \leq 1\), the smaller the time value is, the safer it is. When \(NHRI > 1\), the non-carcinogenic heavy metals have been contaminated. When \(NHRI > 1\), the higher the time value is, the greater the health risk is.

(2) Construction of health risk assessment model for carcinogenic heavy metals. Four kinds of heavy metal pollution \(C_r, A_r, C_d, N_i\) in the sludge of sewage treatment plant in industrial park have carcinogenic characteristics. The health risk assessment model of carcinogenic heavy metal is constructed by using the EPA human exposure assessment model for reference. Among the four carcinogenic heavy metals in sludge, only \(A_r\) can cause health risk to the contact person by breathing inhalation, hand mouth intake and skin contact, and the other three carcinogenic heavy metals can only cause carcinogenic health risk by breathing inhalation. To study the effects of carcinogenic heavy metal pollution on human health risk, we need to study the life cycle of the receptor. Considering that the average life span of Nanjing, China is 79 years, then the average exposure time needs to be scientifically determined. If \(LD_{\text{DE}}\) represents the daily average exposure dose (mg/kg·d) over the life cycle of a person exposed to carcinogenic heavy metals through inhalation, and
$CAT$ represents the average exposure time (d) for carcinogenic heavy metals, the average exposure time of a full cycle can be expressed as the product of the average lifetime of the heavy metal exposure site and the number of days exposed in a year. The standards of cancer risk caused by carcinogenic heavy metals in children and adults are different. $LIR_{\text{child}}$ and $LIR_{\text{adult}}$ represent the respiration rate (m$^3$/d) of heavy metals contamination in children and adults respectively. $LYE_{\text{child}}$ and $LYE_{\text{adult}}$ represent the years of exposure to heavy metal contamination in children and adults (a) respectively. $AW_{\text{child}}$ and $AW_{\text{adult}}$ represent the average weight of children and adults respectively.

\[
LDED_{ij} = \frac{C \cdot EF \left( LIR_{\text{child}} \cdot LYE_{\text{child}} \cdot AW_{\text{adult}} + LIR_{\text{adult}} \cdot LYE_{\text{adult}} \cdot AW_{\text{child}} \right)}{PEF \cdot CAT \cdot AW_{\text{child}} \cdot AW_{\text{adult}}}
\]  

(12)

The above formula is the whole life cycle daily exposure dose measurement model of carcinogenic heavy metals in sludge of industrial park sewage treatment plants through respiratory pathway. Only $A_s$ among the carcinogenic heavy metals can produce carcinogenic risk through hand mouth ingestion and skin contact, and the other three carcinogenic heavy metals do not produce carcinogenic risk through hand mouth ingestion and skin contact. Therefore, the daily average exposure dose measurement model of other carcinogenic heavy metal life cycle can be simplified. In the respiratory intake pathway, there is: $LIR = NRR$; in the hand mouth intake pathway, there is: $LIR = IR$; in the skin contact pathway, there is: $LIR = ESA \cdot SA \cdot SAF$. The health risk of carcinogenic heavy metals in the sludge of sewage treatment plant in industrial park has the single factor carcinogenic health risk of some heavy metal and the multi factor carcinogenic health risk of many heavy metals. When $SCR_{ij} = LDED \cdot SF_{ij}$ is used to indicate the carcinogenic risk of $i$th heavy metal in the $j$th ingestion pathway, $SF_{ij}$ is a carcinogenic factor, reflecting the maximum probability of carcinogenesis of the $i$th heavy metal in the $j$th contaminated ingestion mode. $CCR_{ij}$ represents the combined carcinogenic risk of all carcinogenic heavy metals in the sludge under all ingestion patterns. The corresponding carcinogenic risk model is:

\[
CCR_{ij} = \sum_{j=1}^{m} \sum_{i=1}^{n} SCR_{ij} = \sum_{j=1}^{m} \sum_{i=1}^{n} LDED \cdot SF_{ij}
\]  

(13)
The above is the health risk assessment model of carcinogenic heavy metals in sludge of sewage treatment plant in industrial park. In the health risk assessment of single carcinogenic heavy metals, the health risk assessment criteria of non-carcinogenic heavy metals are detailed in the reference agent scale. The comprehensive health risk assessment criteria for all carcinogenic heavy metals, based on national standards and existing research results, were determined as: no risk of carcinogenesis when $CCR < 10^{-6}$. It is considered that the standard is micro-pollution when $CCR < 10^{-4}$, the risk of carcinogenesis is extremely low. When $CCR \geq 10^{-4}$, it is considered as carcinogenic risk, the greater $CCR$ is, the greater risk of carcinogenesis is.

### 2.2.3 Model parameter determination and assessment standard

In order to assess the health risk of heavy metals in sludge of sewage treatment plant in industrial park, the parameters of the above construction assessment model should be determined, and the value of the evaluation standard should be given according to the requirements of health risk assessment. This work includes the following elements:

1. Determination of Nemerow Index Method parameters and standard. In this method, the evaluation standard value of heavy metal content in sludge $S_i$ is mainly determined. This standard has been changed from mandatory to recommended since March 2017, according to China's *Urban Sewage Treatment Sludge* (GB T24188-2009). The main control of sludge in the sewage treatment plant of Nanjing MV Industrial Park includes eight kinds of heavy metals: Zn, Cu, Pb, Hg, Cr, Ni, As, Cd. The corresponding limit content evaluation standard values of heavy metals are: 4000 mg/kg, 1500 mg/kg, 1000 mg/kg, 25 mg/kg, 1000 mg/kg, 200 mg/kg, 75 mg/kg, 20 mg/kg. However, this standard in the evaluation is obviously too low to truly reflect the characteristics of heavy metal pollution in sludge of sewage treatment plants in industrial parks. To facilitate the application of the evaluation results, referring to *China Soil Environmental Quality Standard* (GB15618-1995) and using the natural background level 1 standard, the corresponding standard are
determined respectively as follows: 100 mg/kg, 35 mg/kg, 90 mg/kg, 40 mg/kg, 15 mg/kg, 35 mg/kg, 0.2 mg/kg, 0.15 mg/kg. By using these standards, it is convenient to use Nemerow Index Method to make a single and comprehensive assessment of the pollution degree of heavy metals in the sludge of sewage treatment plants in industrial parks.

(2) Determination of Muller Index Method parameters and standard. The arithmetic mean original values of geochemistry background values in Nanjing are: 80.71 mg/kg, 33.03 mg/kg, 31.28 mg/kg, 0.0764 mg/kg, 76.95 mg/kg, 36.11 mg/kg, 9.24 mg/kg and 0.202 mg/kg, according to the research results of Wang Hao in Nanjing University. Then, the parameters of heavy metal exposure risk in sludge of industrial park sewage treatment plant are determined, which is also the main parameters in equation (10)–(13). Heavy metal exposure risk parameters were determined by reference to foreign literature (Miguel et al., 2007; Lim et al., 2008). The main parameters meaning of the health risk assessment model of heavy metal pollution in sludge of sewage treatment plants in industrial parks and the evaluation standard value are arranged as follows:

| Serial number | Parameter symbol | Parameter name | Unit | Health risk standard values |
|---------------|-----------------|----------------|------|-----------------------------|
| 1             | NRR             | Respiratory frequency | m³/d | 7.65 (Children) 12.86 (Adult) |
| 2             | IR              | Frequency of ingestion through hand | mg/d | 250 (Children) 150 (Adult) |
| 3             | YE              | Years of exposure | d | 8 (Children) 25 (Adult) |
| 4             | AE              | Exposure frequency | d/a | 320 (Children) 350 (Adult) |
| 5             | NAT             | Average non-carcinogenic exposure time | d | 20 (Adult) |
| 6             | CAT             | Average carcinogenic exposure time | d | 25 (Adult) |
| 7             | AW              | Average weight | kg | 16 (Children) 65 (Adult) |
| 8             | PEF             | Particulate matter emission factor | m³/d | 1.36E9 (Children) 1.36E9 (Adult) |
| 9             | ESA             | Exposed skin surface | cm² | 2000 (Children) 5000 (Adult) |
| 10            | SA              | Skin adhesion | mg/(cm²)(d) | 0.2 (Children) 0.07 (Adult) |

The characteristics of heavy metal pollution and the main parameters of the health risk assessment model in the sludge of sewage treatment plant in industrial park are determined above. In order to effectively assess the environmental pollution status and the size of health risk, it is also necessary to determine the health risk assessment standard of non-carcinogenic and carcinogenic heavy metals under different ingestion
pathways according to the assessment requirements. The results are detailed in Table 6.

Table 6 Non-carcinogenic reference dose and carcinogenic slope factor of heavy metals in sludge

| Category       | Heavy metals | Non-carcinogenic heavy metal reference dose (RfD) (mg/m³·d) | Hand and mouth intake (mg/kg·d) | Exposure to skin (mg/m²·d) |
|----------------|--------------|----------------------------------------------------------|---------------------------------|---------------------------|
| Non-carcinogenic risk criteria | P̃            | 3.52E-2                                                  | 3.50E-3                         | 5.25E-4                   |
|                | Zn           | 0.300                                                   | 3.00E-1                         | 6.00E-2                   |
|                | Cu           | 4.02E-2                                                  | 4.00E-2                         | 1.20E-2                   |
|                | Hg           | 9.00E-5                                                  | 3.00E-4                         | 2.00E-5                   |
| Cancer risk standard | Cr          | 42                                                      | --                              | --                        |
|                | As           | 15.10                                                   | 1.50                            | 3.66                      |
|                | Cd           | 6.30                                                    | --                              | --                        |
|                | Ni           | 0.84                                                    | --                              | --                        |

3. Results and Discussions

3.1 Characteristics assessment of heavy metal pollution in sludge

Nanjing MV Industrial Park is located along the Yangtze River in the Pukou High-tech Development Zone in Nanjing, China, with an area of 52.98 square kilometers. There are 148 production enterprises in the park. In order to efficiently treat the sewage discharge produced by the enterprises in the park, the sewage treatment funded by the government was built, which has a wastewater treatment capacity of 22,000 M³perday. About 16,500 tons of sludge is produced annually at the large sewage treatment plant. Sludge is mostly produced by industrial wastewater treatment, so the enterprises in the park are mostly chemical enterprises. The sewage treatment plants monitor and control eight heavy metals contained in the sludge, including Zn, P̃, Cu, Cd, Cr, Ni, Hg and As. According to the research design, the characteristics of heavy metal pollution in sludge of sewage treatment plant in Nanjing MV industrial park are comprehensively evaluated.

3.1.1 Evaluation results of Nemerow Index Method

Nemerow IndexMethod is a classical pollution condition evaluation method, which is relatively simple. In this study, the relative of the evaluation index is added to the comprehensive index calculation of Nemerow
Index Method, which is used to improve the evaluation effect. The evaluation standard refers to the soil environmental quality standard.

Table 7 Nemrow Index Method Evaluation Results of Heavy Metal Pollution Characteristics

| Serial number | Heavy metals | Contamination Standards (mg/kg) | Single pollution index | Relative Weight | Weighted value | Pollution level |
|---------------|--------------|---------------------------------|------------------------|----------------|----------------|----------------|
| 1             | Pb           | 34.73                           | 35                     | 300            | 0.9923         | 0.1158         | I              | I              |
| 2             | Zn           | 100.67                          | 100                    | 250            | 1.0067         | 0.4027         | I              | I              |
| 3             | Cu           | 50.64                           | 35                     | 100            | 1.4469         | 0.5064         | I              | I              |
| 4             | Hg           | 0.18                            | 0.15                   | 0.5            | 1.2000         | 0.3600         | I              | I              |
| 5             | Cr           | 58.37                           | 90                     | 300            | 0.6486         | 0.1946         | I              | I              |
| 6             | As           | 16.41                           | 15                     | 25             | 1.0940         | 0.6564         | II             | I              |
| 7             | Ni           | 38.64                           | 40                     | 50             | 0.9660         | 0.7728         | II             | I              |

Total pollution index = \( \sum \text{relative weights} \times \text{individual pollution index} \)

According to the above calculation results, the pollution status of heavy metals is ranked in order of heavy metal pollution index as: \( C_\text{a} > H_g > C_d > A_s > Z_n > P_b > N_i > C_r \). Thus, using lower evaluation standard, heavy metals in sludge of most treatment plants in Nanjing MV industrial park have safety characteristics. If the evaluation standards are improved, the evaluation results show that the three heavy metals Cu, As, Ni in sludge have the characteristics of micro degree risk, and the comprehensive condition of heavy metals in sludge also presents micro degree risk characteristics. It can be seen that the evaluation results of this evaluation method are loose.

3.1.2 Evaluation results of Muller Index Method

Muller Index Method, which is also called Geoaccumulation index Method, judges the pollution condition by multiplying the measured value and the geochemical background value of the sludge containing a certain heavy metal by the logarithm geochemical background the product ratio before industry. The earth background value \( k \) of Nanjing refers to the research results of Wang Hao of Nanjing University. Pre-Industrial
geochemical background $B_i$ refers to the research results of Li Qingfang et al. The Muller Index Method evaluation results of heavy metal pollution characteristics in sludge of sewage treatment plants of Nanjing MV industrial park are detailed in Table 8 according to the above research design.

| Serial number | Heavy metals | Heavy metal content (mg/kg) | $k$ | $B_i$ | $I_{geo}$ | Pollution level |
|---------------|--------------|-----------------------------|-----|-------|----------|-----------------|
| 1             | $Pb$         | 34.73                       | 80.71 | 26    | -2.6508  | I               |
| 2             | $Zn$         | 100.67                      | 33.03 | 80    | 0.3306   | II              |
| 3             | $Cu$         | 50.64                       | 31.28 | 26    | 1.4816   | III             |
| 4             | $Hg$         | 0.18                        | 0.0764 | 0.06 | 0.6894   | II              |
| 5             | $Cr$         | 58.37                       | 76.95 | 80    | -1.2735  | I               |
| 6             | $As$         | 16.41                       | 36.00 | 5     | 1.3618   | III             |
| 7             | $Cd$         | 0.22                        | 9.24  | 0.11  | -0.4729  | I               |
| 8             | $Ni$         | 38.64                       | 0.202 | 32    | 1.0236   | III             |

According to the above evaluation results, it is obvious that the heavy metal pollution in sludge of sewage treatment plants in Nanjing MV Industrial Park has been more serious by using this evaluation method. The pollution level of heavy metals $Cu$, $As$, $Ni$ have raised to level III and that of heavy metal $Zn$ and $Hg$ has risen to level II.

3.1.3 Evaluation results of Grey Clustering Method

In order to realize the comprehensive evaluation of heavy metal pollution characteristics in sludge of sewage treatment plants in industrial parks by using Grey Clustering Method, the relative weights of the evaluation index need to be determined first. According to the measured results of heavy metals in sludge of sewage treatment plants in Nanjing MV Industrial Park in Table 1, the data in the table is $x_{ij}$. For saving space, the specific original data matrix expression is omitted. The original data matrix is standardized by using formula (11) to obtain the standardized matrix. According to the above standardized matrix, the entropy of the evaluation index is calculated by using formula (13). After the ratio of entropy $f_{ij}$ is translated by formula (14), the relative weight value of the evaluation index is calculated by using formula (15) as follows:

$$\xi_{ij} = (0.1452, 0.0814, 0.0752, 0.2295, 0.0878, 0.1118, 0.1695, 0.0996)$$
After determining the grey class of grey cluster evaluation, the evaluation standard of grey class is set. Using the measured data of heavy metal content in sludge samples of sewage treatment plant in Nanjing MV Industrial Park and the membership function (whitening weight function of gray number), the membership degree of evaluation index is calculated for the standardized measured data of heavy metal content, the index variable weight clustering coefficient of grey evaluation grade $K$ can be determined by the product of the evaluation index membership matrix and the evaluation index relative weight matrix. Grey Clustering Method evaluation results of heavy metal pollution characteristic and two other pollution characteristic evaluation results can be determined in Table 9.

| Heavy metals | Measured contents of heavy metals | Relative weights of evaluation indicators | Gray Class $k$ | Gray class corresponding to maximum element |
|--------------|-----------------------------------|------------------------------------------|----------------|-------------------------------------------|
| $Pb$         | 34.73                             | 0.1452                                   | 0.2552         | Ⅰ                                          |
| $Zn$         | 100.67                            | 0.0814                                   | 0.2461         | Ⅰ                                          |
| $Cu$         | 50.64                             | 0.0752                                   | 0.2438         | Ⅱ                                          |
| $Hg$         | 0.18                              | 0.2295                                   | 0.2538         | Ⅱ                                          |
| $Cr$         | 58.37                             | 0.0878                                   | 0.2638         | Ⅰ                                          |
| $As$         | 16.41                             | 0.1118                                   | 0.2547         | Ⅱ                                          |
| $Cd$         | 0.22                              | 0.1695                                   | 0.2638         | Ⅰ                                          |
| $Ni$         | 38.64                             | 0.0996                                   | 0.2637         | Ⅱ                                          |

According to the grey clustering evaluation method, in the results of the evaluation in the table above, the category of heavy metal pollution features was determined according to the maximum meta-value principle and the results of the evaluation of the Grey Clustering Method in action 9. Compared with the evaluation results of Nemerow Index Method and Muller Index Method, the evaluation results of grey clustering evaluation method are located in the middle of the evaluation results of the other two heavy metal pollution characteristics, and have the attribute of taking the best from the worst. Hence, using Grey Clustering Method to evaluate heavy metal pollution characteristics in sludge of sewage treatment plants in Nanjing MV industrial park has a good evaluation effect.
3.2 Health risk assessment of heavy metal pollution in sludge

3.2.1 Health risk assessment of non-carcinogenic heavy metals

Among the non-carcinogenic heavy metals in sludge of sewage treatment plant of Nanjing MV Industrial Park, there are four main heavy metals including Pb, Zn, Cu, Hg. According to the above research design and the measured results of heavy metal content in sludge samples obtained in this study, the health risk of non-carcinogenic heavy metals in sludge of sewage treatment plant of Nanjing MV Industrial Park is comprehensively evaluated, and the specific evaluation results are detailed in Table 10.

According to the above assessment results of health risk of non-carcinogenic heavy metals in sludge of sewage treatment plant in industrial park, the order of health risk impact of non-carcinogenic heavy metals on children and adults is: Hg, Cu, Pb, Zn, and the order of health risk impact of the three routes of heavy metal exposure was: hand mouth intake, skin contact and respiration. The health risk of non-carcinogenic heavy metal pollution in children was significantly higher than that in adults, and nearly twice as high as that in adults, which indicated that children had higher non-carcinogenic heavy metal pollution tolerance than adults.

3.2.2 Health risk assessment of carcinogenic heavy metals

Among the carcinogenic heavy metals in sludge of the sewage treatment plant in Nanjing MV Industrial
Park, there are mainly four kinds of heavy metals, including \( C_{d} \), \( A_{r} \), \( C_{d} \), \( N_{i} \). According to the above research design and the measured results of heavy metal content in sludge samples obtained in this study, the health risk of carcinogenic heavy metals in sludge of sewage treatment plants in Nanjing MV Industrial Park is comprehensively evaluated, and the specific evaluation results are detailed in Table 11.

Table 11: Health risk assessment of carcinogenic heavy metals

| Heavy metals | \( C_{r} \) | \( A_{r} \) | \( C_{d} \) | \( N_{i} \) | Total |
|--------------|----------------|----------------|----------------|----------------|----------------|
| \( BI \)     | 3.54E-06       | 3.52E-06       | 3.12E-04       | 6.52E-6        | --             |
| \( HMI \)    | 2.98E-05       | 2.67E-04       | 3.54E-04       | 4.35E-05       | --             |
| \( SC \)     | 5.46E-06       | 3.45E-04       | 3.36E-05       | 5.82E-05       | --             |
| \( BI \)     | 4.85E-06       | 3.42E-05       | 4.92E-04       | 6.57E-06       | --             |
| \( HMI \)    | 3.55E-05       | 4.87E-04       | 4.35E-04       | 3.85E-05       | --             |
| \( SC \)     | 6.12E-06       | 4.58E-04       | 4.87E-05       | 4.87E-06       | --             |
| \( BI \)     | 2.521E-05      | 5.87E-06       | 2.35E-05       | 3.47E-05       | 8.93E-05       |
| \( HMI \)    | --             | --             | 3.46E-04       | --             | 3.46E-05       |
| \( SC \)     | --             | --             | 2.66E-04       | --             | 2.66E-04       |
| \( \Sigma \) | 2.521E-05      | 5.87E-08       | 3.96E-05       | 3.47E-05       | --             |
| \( BI \)     | 4.221E-05      | 6.85E-06       | 3.45E-05       | 3.76E-05       | 1.21E-04       |
| \( HMI \)    | --             | --             | 3.26E-04       | --             | 3.26E-04       |
| \( SC \)     | --             | --             | 3.85E-04       | --             | 3.85E-04       |
| \( \Sigma \) | 4.221E-05      | 6.85E-06       | 7.46E-04       | 3.76E-05       | --             |

According to the assessment results of the health risks of carcinogenic heavy metals in sludge of the sewage treatment plants in industrial parks above, the order of health risk effects of carcinogenic heavy metals on children and adults is: \( C_{d}, C_{r}, N_{i}, A_{r} \), and the order of health risk impact of the three ways of heavy metal exposure was: skin contact, hand intake and respiratory inhalation. The health risk tolerance of carcinogenic heavy metal pollution in children is slightly lower than that of adults, which indicates that carcinogenic heavy metal pollution is more likely to cause carcinogenic harm to children.

3.3 Discussion of Assessment Results

1. Comparison of results and selection of methods for assessment of pollution characteristics. In order to
make an effective assessment of heavy metal pollution characteristics of sludge of sewage treatment plant in Nanjing MV Industrial Park, we selected three pollution feature assessment methods: Nemerow Index Method, Muller Index Method and Grey Clustering Method. The first two assessment methods are classical pollution characteristics assessment methods, and the latter one is a pollution characteristics assessment method introduced in this paper. According to the evaluation results of three methods, the cone diagram is used to represent the level of heavy metal pollution grade in sludge of sewage treatment plants in Nanjing MV Industrial Park. According to the research design, the pollution grade of heavy metals is divided into five levels: pollution-free, micro pollution, light pollution, moderate pollution, heavy pollution, corresponding to levels one through five. The results and trends of heavy metal pollution level assessment by three methods are shown in Figure 1.

From the above image, it can be seen that the characteristics of heavy metal pollution in sewage treatment plant in Nanjing MV Industrial Park show a low pollution level, and the evaluation results of different methods are limited to the level of light pollution. Nevertheless, different assessment methods have great influence on the evaluation results. Generally speaking, the pollution characteristic grade of Nemerow Index Method assessment is relatively low, the pollution characteristic grade of Muller Index Method assessment is relatively high, and the pollution characteristic grade of Grey Clustering Method assessment is
located between the evaluation results of the other two methods. The sewage treatment plant in industrial park can choose different assessment methods according to the requirements and objectives of heavy metal characteristics assessment in sludge. Comparison of the results of the three different methods also reveals that: the heavy metal pollution grades of Cu, As and Ni in sludge of sewage treatment plant in Nanjing MV Industrial Park are relatively high, the heavy metal pollution grades of Pb, Cr and Cd are relatively low, and the heavy metal pollution grades of Hg and Zn are at the middle level.

(2) Health risk assessment findings and cause analysis. Through the above research, it is found that Hg and Cu has the greatest impact on health risk among non-carcinogenic heavy metal pollution in sewage treatment plant of Nanjing MV Industrial Park, Cd and Cr has the greatest impact on health risk among carcinogenic heavy metal pollution. The health risk of non-carcinogenic heavy metal pollution in children is much higher than that in adults. The health risk of carcinogenic heavy metal pollution in children is slightly lower than that in adults. The impact of heavy metals on human health risk is mainly through exposure. These conclusions are obtained by the analysis of the results of the study and can be reflected by Figure 2.

![Figure 2 Composition and Trends of Health Risk Assessment Results](image_url)

According to the above research findings, the main causes of its production are revealed on the basis of comprehensive analysis. There are two reasons for the high content of heavy metal Cu and Hg in sludge, one is that the content of these two heavy metals in industrial wastewater itself is high, and the content of sewage
treatment residue in sludge is high; the other is that the current results are due to the lack of control measures and removal technology during sewage treatment, which need to be reduced by taking technical measures according to the evaluation results. The health risk of non-carcinogenic heavy metal pollution in children is greater than that in adults, which is the result of normative research. The main reason for the analysis is that children are in the growth period, and the future resistance is stronger than the current resistance. Also, because children are far away from sludge, exposure to and ingestion of heavy metals are much less likely than adults. However, the effects of carcinogenic heavy metals on health risks are less likely to cause harm to the human body, so children with lower physical fitness are less likely to be able to accept it. The health risk of heavy metal is mainly due to the presence of heavy metals in the sludge, the exposure time and the distance from the sludge. The people exposed to the sludge in the industrial park are all adults, the exposure to heavy metals must be more, and the health risk of heavy metal field and those who are far away from the sludge site is naturally small.

4. Conclusions

The research topic of heavy metal pollution characteristics and health risk assessment in sludge of sewage treatment plants in industrial parks is very complex. To explore the characteristics of heavy metal pollution and the effective methods of health risk assessment in sludge of sewage treatment plants in China industrial parks, three research methods were selected, including Nemerow Index Method, Muller Index Method and Grey Clustering Method based on literature review and current situation analysis. The pollution characteristics of eight heavy metals (Zn, Cu, Pb, Hg, Cr, Ni, As, Cd) in sludge of sewage treatment plants in Nanjing MV industrial park were evaluated and applied. On this basis, the health risk assessment of four non-carcinogenic heavy metals Pb, Zn, Cu, Hg and four carcinogenic heavy metals Cr, As, Cd, Ni was studied. In addition to distinguishing heavy metals into non-carcinogenic heavy metal pollution health risk assessment and carcinogenic heavy metal pollution health risk assessment, the health risk assessment of
children and adults and its application in two heavy metal states were also studied. According to the research results, in the pollution characteristics assessment study of heavy metal in sludge, the research method has a great impact on the heavy metal pollution characteristics assessment results, the heavy metal pollution grade of Cu, As and Ni is relatively high among all the heavy metals. Therefore, it is very important to choose a reasonable assessment method, the different research methods will make the order of heavy metal pollution impact change, and controlling focus is also an effective strategy. In the assessment of non-carcinogenic heavy metal health risk, Hg and Cu have the greatest impact on health risk, children have a higher tolerance to health risk from non-carcinogenic heavy metal pollution than adults, children have higher health risk tolerance than adults, and the corresponding tolerance is nearly twice as high. In the assessment of carcinogenic heavy metal health risk, heavy metal Cd and Cr have the highest carcinogenic risk, adult's ability to bear the health risk of carcinogenic heavy metal pollution is slightly higher than children's. In the three pathways of heavy metal intake into the human body, the most hazardous way is skin exposure to heavy metals, the second is the hand intake of heavy metals, and the third is breathing inhalation of heavy metals. Therefore, reducing the contact with sludge is the key to reduce the carcinogenic risk of heavy metals in sludge of sewage treatment plants.

**Abbreviations**

MV: The name of the target company is not authorized to be expressed in letters; NIM: Nemerow Index Method; MIM: Muller Index Method; GCM: Grey Clustering Method.

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**Authors’ contributions**

Under the guidance of Professor Sun Tao, this paper was written by Dr. Xiuyan Han and translated into English by Ms. Tianyi Cao. Professor Tao Sun designed the paper and participated in the modeling. Ms. Tianyi Cao also participated in the technical processing of research data and related calculations. The results of this
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**Availability of data and materials**

The data and materials in this paper are included in the paper and the annex.

**Ethics approval and consent to participate**

Not applicable, since the present study is a re-evaluation of existing data.

**Consent for publication**

Not applicable

**Competing interests**

Professor Tao Sun is employed by Nanjing University of Aeronautics and Astronautics. Xiuyan Han is a doctoral student and Tianyi Cao is a master. This paper is the research result of Professor Tao Sun's project of China Social Science Foundation. Therefore, there is no competitive interest between this article and any person or organization.

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