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Comprehensive evaluation of leaching toxicity of tailings in non-resource-based cities in Anhui Province based on entropy weight-clustering method

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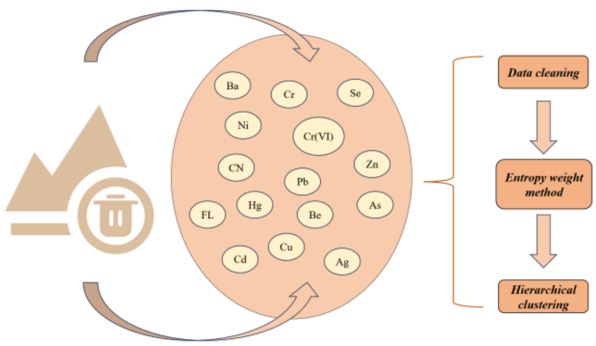
Graphical abstract



Abstract

Toxic components in tailings pose a severe threat to the ecosystem and human health and negatively affect tailings' resource utilization. Therefore, using the entropy weight-clustering method, we evaluated the leaching toxicity based on tailings samples from non-resource cities in Anhui Province. The results show that, all the extracted tailings samples first passed the leaching toxicity test, but there is still a large room for improvement. Secondly, there are complex interrelationships among the 15 indicators measuring leaching toxicity. Thirdly, in the process of entropy weight method calculation, the top three indicators in terms of weight are Cu (17.65%), Ag (16.52%), and Be (8.66%). Finally, the Ward clustering results show that the toxicity scores of tailings in each mining area exhibit distinct hierarchical characteristics, and they can be divided into two major categories and three subcategories (I-1, I-2, II), with the toxicity scores of category I-1 significantly higher than the rest of the mining areas. These findings provide empirical evidence for targeted treatment of tailings resources.

Keywords: tailings toxicity, entropy weighting method, hierarchical clustering, non-resource cities

1. Introduction

It is widely acknowledged that humans have had an indelible impact on planetary processes and that human activity has changed the earth enough to usher in a new geological era, the Anthropocene (Malhi 2017; Waters *et al.* 2016; Zeng *et al.* 2020). In the context of the Anthropocene and post-covid-19 era, depletion of mineral resources, environmental degradation, and natural resource exploitation are the three significant global challenges (Xiong *et al.* 2023), and the resource utilization of tailings is a synergistic and effective way to address these three challenges.

Tailings are "waste" slag left after extracting useful substances or elements from raw ore from metal or non-metal mines. The rapid mining of mineral resources is accompanied by massive tailings (Guo *et al.* 2022). Since reform and opening up, China's rapid urban development and economic growth have led to an expanding demand for mineral resources (Liu & McDonald 2010; Shen *et al.* 2005), resulting in a concomitant increase in tailings emissions. According to statistics, in 2018, the tailings generation of key survey industrial enterprises in China was 880 million tons, accounting for 27.4% of the general solid waste generation of key survey industrial enterprises. However, its comprehensive utilization rate was only 27.1%, which is still a big gap with the average comprehensive utilization rate of other industrial solid wastes (Wang *et al.* 2020). One of the critical factors affecting the resource utilization of tailings is their toxic composition.

Generally, tailings contain high concentrations of toxic metals and metalloids (Nguyen *et al.* 2021). These toxic components in tailings pose a severe threat to the ecosystem and human health and negatively affect the resource utilization of tailings. High concentrations of toxic elements in tailings can make it particularly difficult to utilize them, as treatment of such elements often requires complex scientific techniques and high economic costs. In addition, the tailings pond is not only a pollution source, but also a risk source for safety accidents. Recent studies have shown that microorganisms also have an impact on the safety of tailings (Cao *et al.* 2019; Wang *et al.* 2022).

Current methods for assessing the toxicity of tailings can be categorized into environmental impacts and health risks. The most used assessment methods in the environmental impact category are the geoaccumulation index (Igeo) proposed by Muller (1969) and the potential ecological risk assessment (RI) proposed by Hakanson (1980). Among them, Igeo provides a basis for assessing soil contamination in mining areas by comparing the metal concentrations found in tailings samples with those of geochemical background value (Li *et al.* 2014).RI, on the other hand, is based on a specific set of equations, which can be used to assess the potential ecological risk index by placing the metal concentrations of tailings samples, the corresponding background value, and the toxic biological response factor as variables(Lima *et al.* 2024; Shen *et al.* 2019). Unlike environmental impact assessment methods, health risk assessment for tailings toxicity requires multiple steps. First, an average daily intake (ADI) is calculated based on the three exposure pathways: oral ingestion, dermal contact, and inhalation. Second, the hazard quotient (HQ) is calculated based on the reference dose (RfD) with ADI, which assesses the non-carcinogenic risk. Finally, the carcinogenic risk (CR) is calculated based on the slope factor (SF) with ADI (Kan *et al.* 2021). By sorting out the toxicity assessment methods of tailings, it can be found that most of the current commonly used assessment methods need to rely on the background value with parameters, which may lead to measurement errors. The entropy value method, as a commonly used objective assignment method, can effectively reduce the errors caused by improper parameter setting and has been widely used in the assessment of coal mine safety and green mine construction level in recent years (Li *et al.* 2011; Yin *et al.* 2024). Therefore, we have developed a new comprehensive framework for assessing the leaching toxicity of tailings and conducted analysis based on tailings samples from Anhui Province, which provides empirical evidence for targeted treatment of tailings resources.

The rest of the paper is structured as follows. Section 2 outlines the sample selection and analysis process, providing a brief overview of the statistical methods used in this paper. Section 3 presents the results, including descriptive statistics, correlation analysis, entropy weight analysis, and clustering analysis. Finally, in section 4, the findings are discussed and summarized.

2. Materials and methods

2.1. Sample selection

The study area of this paper is Anhui Province in China. Located in the central-eastern part of China, Anhui Province boasts diverse and complex geographical features, predominantly characterized by hills and mountains. It is a significant base for China's energy, raw materials, and manufacturing (Jiang *et al.* 2018). Mineral resources in Anhui Province are relatively wealthy, and many mines exist. As of the end of 2015, 76 kinds of solid minerals (including subspecies) were utilized in Anhui Province, with 1638 mines, including 835 production mines, accounting for 50.98%. Solid mines produced 465 million tons of ore annually, with an output value of 77.6 billion yuan, accounting for 3.53% of the province's gross domestic product (Bureau of Geology and Mineral Exploration of Anhui Province 2018). We selected three mining areas in Huoqiu County, Anhui Province (designated as KF, LT, and FQ), and two mining areas in Lujiang County, Anhui Province (designated as JD and WX) as sampling sites. This choice was made because these two regions are neither resource-based cities nor resource-depleted areas, thus exhibiting typicality and representativeness regarding natural resource endowment. In addition, this sample selection approach also facilitates extending the research experience to other regions, which can make a more outstanding theoretical contribution to the resource utilization of tailings.

2.2. Sample analysis

Twenty-six samples from five mining areas were analyzed for leaching toxicity by the *Solid waste-Extraction procedure for leaching toxicity-Sulphuric acid & nitric acid method* (HJ/T 299-2007 2007). Final measurements include Copper (Cu), Zinc (Zn), Lead (Pb), Chromium (Cr), Hexavalent Chromium (Cr(VI)), Beryllium (Be), Barium (Ba), Cadmium (Cd), Nickel (Ni), Silver (Ag), Arsenic (As), Selenium (Se), Mercury (Hg), Fluoride (FL), and Cyanide (CN). All 15 indicators will be used to assess the leaching toxicity of tailings.

2.3. Evaluation criteria

We chose to evaluate the leaching toxicity of tailings at each mine site using the entropy weight method. The basic ideology of entropy weight theory is that if the values of a given indicator differ more between the evaluation samples, then it means that the indicator provides more information. When an indicator is equal in the evaluation sample, it is automatically filtered in the calculation process. In other words, the weight of such an indicator will be zero (Zhao *et al.* 2018). Drawing on Amiri *et al.* (2014), the entropy weight method is applied in this paper in the following steps:

The first step is the dimensionless of the indicators. Because this study only deals with negative indicators, it is only necessary to use normalization for negative indicators:

$$x_{ij}^{'} = \frac{max\, x_{ij} - x_{ij}}{max\, x_{ij} - min\, x_{ij}} \tag{1}$$

The second step calculates the ratio p_{ij} of the i evaluation object on the j indicator:

$$p_{ij} = \frac{x_{ij}^{'}}{\sum_{i=1}^{m} x_{ij}^{'}} \tag{2}$$

The third step calculates the entropy value e_j for each indicator based on p_{ij}:

To eliminate the effect of $p_{ij} = 0$ in equation (3), we apply a non-negative translation of 0.001 units.

The fourth step calculates the weights w_j for each indicator:

$$e_j = -\frac{1}{\ln m}\sum_{i=1}^{m} p_{ij}\ln\left(p_{ij}\right) \tag{3}$$

$$w_j = \frac{1-e_j}{\sum_{j=1}^{n}\left(1-e_j\right)} \tag{4}$$

Finally, the toxicity score z_i is calculated from the normalized data $x_{ij}^{'}$ with weights w_j:

$$z_i = \sum_{j=1}^{n} x_{ij}^{'} \times w_j \tag{5}$$

After calculating the comprehensive scores for the leaching toxicity of tailings in each mining area, we used the Ward hierarchical clustering method for classification. The Ward method, also known as the Ward variance minimization algorithm, initially considers each sample in the population as a cluster. Then, at each step of merging clusters, it selects the two clusters with the smallest increase in the sum of squared deviations and merges them until all samples are grouped into one cluster (Yang 2010). The specific steps are as follows:

Divide *n* samples into *k* clusters: G_1, G_2...G_k. Use $\overline{y_i}$ to denote the centroid of G_i (the mean of samples in this cluster), y_{ij} to represent the *j* sample in G_i, and n_i to denote the number of samples in G_i. Then, the sum of squared deviations of samples in G_i is:

$$S_i = \sum_{j=1}^{n}\left(y_{ij}-\overline{y_i}\right)^{'}\left(y_{ij}-\overline{y_i}\right) \tag{6}$$

The sum of squared deviations within each of the *k* clusters is:

$$S' = \sum_{i=1}^{k}\sum_{j=1}^{n_i}\left(y_{ij}-\overline{y_i}\right)^{'}\left(y_{ij}-\overline{y_i}\right) \tag{7}$$

3. Results

Table 1 shows the descriptive statistics of the leaching toxicity of the samples. Table 1 demonstrates that the concentrations of all detection factors of the selected samples are lower than the thresholds of the Identification standards for hazardous wastes-Identification for extraction toxicity (GB5085.3-2007 2007). It should be noted that the standard deviation of most of the indicators is significant, indicating a large degree of dispersion of the samples. In other words, although the leaching toxicity of tailings samples meets the basic requirements, there is still much room for improvement (Figure 1).

Figure 2 visually represents the correlations between the 15 indicators measuring leaching toxicity. From Figure 2, it can be observed that there are complex relationships among these 15 indicators. Using only one or a few indicators as surrogate variables for tailings toxicity would result in losing crucial decision-making information. Therefore, to systematically evaluate the potential toxicity of Anhui Province tailings, we used the entropy weight method to assess the 15 indicators comprehensively.

According to the previous application steps, we first normalized the 15 indicators. Then, we calculated each indicator's entropy value and weight based on the processed data, and the results are shown in Figure 3.

From Figure 3, the weight of the Cu indicator is the highest, reaching 17.65%, indicating that this indicator provides the most information and has the most significant impact on the evaluation of tailings toxicity. Next, the weight of the Ag indicator also reaches 16.52%, similarly providing rich decision-making information. Furthermore, the weights of the Be, Hg, Ba, As, Se, and Ni indicators are all greater than 5%, indicating that these indicators also significantly influence the comprehensive evaluation of tailings toxicity. Lastly, although the weights of the Zn, FL, Cr, Pb, Cr(VI), Cd, and CN indicators are relatively small, they still threaten ecosystems and human health. Therefore, they should also be taken into consideration when evaluating tailings toxicity.

The toxicity score for each sample can be calculated by substituting the resulting indicator weights into the previous equation (5), and the results are shown in Figure 4. When the mining area is used as the classification standard, it can be found that the fluctuation of the sample scores within the same mining area is relatively smooth, and the scores of each mining area are ranked in the order of WX, FQ, LT, JD, and KF. Among them, the sample scores of the WX mining area are above 0.75, which indicates that it has achieved the best result in the control of tailings toxicity. However, the sample scores for the JD mine, which is in the same domicile as the WX mine, are all within the 0.5-0.75 range, suggesting that the corporate domicile is not a key determinant of its toxicity. Among the three mining areas from Huoqiu County, FQ and LT mining areas achieve relatively good toxicity scores. However, samples from the KF mining area have significantly lower scores than the other mining areas, once again indicating that the domicile determinism cannot adequately explain the toxicity scores of tailings in Anhui Province.

After calculating the toxicity scores for each sample, we further analyzed them using Ward hierarchical clustering with the mine as the basis for classification. Figure 5 is a dendrogram of cluster analysis derived from the automatic partitioning performed using the hierarchical clustering function in the SciPy package of Python. The principles are previously introduced in equations (6) and (7). Specifically, the function's output indicates that the construction of the dendrogram proceeded through four iterations: initially, the distance between the LT and FQ was 0.021, forming the first cluster (I-2); subsequently, the distance between WX and the first cluster was 0.215, merging to form the second cluster (I-1); next, the distance between KF and JD was 0.258, resulting in the third cluster (II); finally, the distance between Cluster I and Cluster II was 0.589, marking the completion of the clustering process. Among them, the I-1 category contains only the WX mine due to a significantly higher toxicity score than the rest of the mines. Secondly, the I-2 category contains the LT and FQ mining areas, and the scores are lower than those of category I-1, but are still at a reasonable level from a global perspective. Finally, the

II category contains the KF and JD mining areas, which have lower scores and more significant potential for improvement.

4. Discussion

The concentrations of the detection factors in the mining samples were all below the thresholds, reflecting that the mining industry has achieved remarkable results as a critical area of solid waste pollution prevention and control. At the same time, the 15 indicators used to measure the toxicity have large standard deviations, indicating that there is still much room for progress in the governance of tailings toxicity in Anhui Province. In further entropy weight method analyses and clustering analyses, we can find that the tailings toxicity scores of the mines show a hierarchical characteristic. The scores of the samples within the same mine are similar, and there is a significant difference in the scores of the samples from different mines. Therefore, it is imperative for mining companies to actively promote innovation in tailings treatment technologies, building upon the achievements of expanding the governance of tailings toxicity. Furthermore, efforts should be intensified to enhance supervision over mining areas with lower scores.

The original contributions of this paper are mainly in terms of the research area and methodology. On the one hand, most of the previous works on tailings in Anhui Province focus on resource-based cities such as Tongling (Yang *et al.* 2014) and Maanshan (Zhou *et al.* 2022). In contrast, this paper takes Lujiang and Huoqiu, two non-resource-based counties as the study area, which is conducive to broadening the understanding of tailings toxicity and making the results more generalizable. On the other hand, due to the complex correlation between the indicators and the hierarchical differences between the samples, this paper chose to employ the entropy weight-clustering method to achieve a comprehensive evaluation of the leaching toxicity of tailings in non-resource-based cities in Anhui Province, which also provides new ideas for academic research in this field.

This paper also had some limitations due to difficulties in data collection. For instance, the sample size used for statistical analyses was only 26, which limited the choice of methodology for this paper. In addition, the small sample size also resulted in the inability to perform refined analysis. Therefore, we hope that future research can expand the sample size and, based on tailings classification (Gou *et al.* 2019), comprehensively evaluate the leaching toxicity of tailings such as silicate minerals, carbonate minerals, feldspar minerals, and clay minerals.

5. Conclusion

In summary, the following conclusions were drawn from this paper. Firstly, all the extracted tailings samples passed the leaching toxicity test, but there is still a large room for improvement. Secondly, there are complex interrelationships among the 15 indicators measuring leaching toxicity. Thirdly, in the process of entropy weight method calculation, the top three indicators in terms of weight are Cu (17.65%), Ag (16.52%), and Be (8.66%). Finally, the Ward clustering results show that the toxicity scores of tailings in each mining area exhibit distinct hierarchical characteristics. They can be divided into two major categories and three subcategories (I-1, I-2, II), with the toxicity scores of category I-1 significantly higher than the rest of the mining areas.

Table 1. Descriptive statistics of toxicity indicators

Variables	Mean	Standard Deviation	Min	Max	GB5085.3-2007	Unit
Cu	0.010	0.009	0.000	0.020	100	mg/L
Zn	0.043	0.058	0.003	0.178	100	
Pb	0.071	0.093	0.001	0.340	5	
Cr	0.054	0.047	0.002	0.170	15	
Cr(VI)	0.005	0.008	0.000	0.032	5	
Be	0.002	0.002	0.000	0.005	0.02	
Ba	0.282	0.410	0.020	1.090	100	
Cd	0.002	0.002	0.000	0.009	1	
Ni	0.034	0.019	0.002	0.070	5	
Ag	0.004	0.005	0.000	0.010	5	
As	0.029	0.041	0.000	0.110	5	
Se	0.111	0.218	0.000	0.550	1	
Hg	0.005	0.008	0.000	0.020	0.1	
FL	0.308	0.166	0.080	0.700	100	
CN	0.025	0.085	0.000	0.399	5	

Note: Round to three decimal places after rounding.

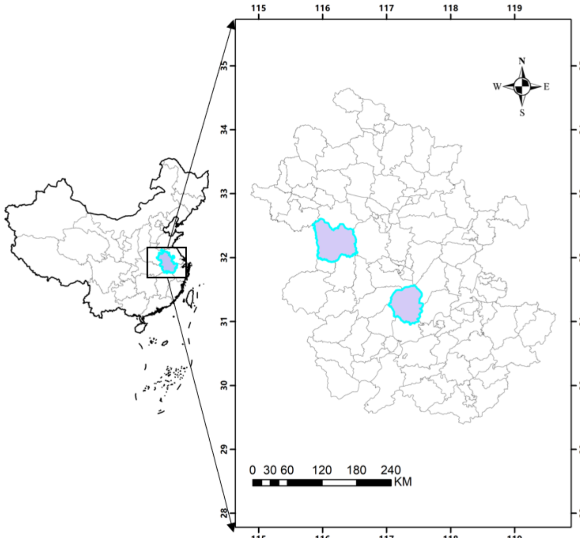


Figure 1. Sampling locations for tailings

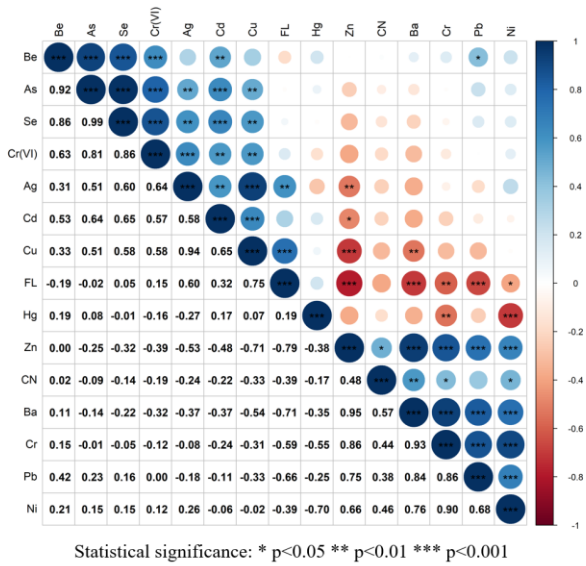


Figure 2. Correlation among toxicity indicators

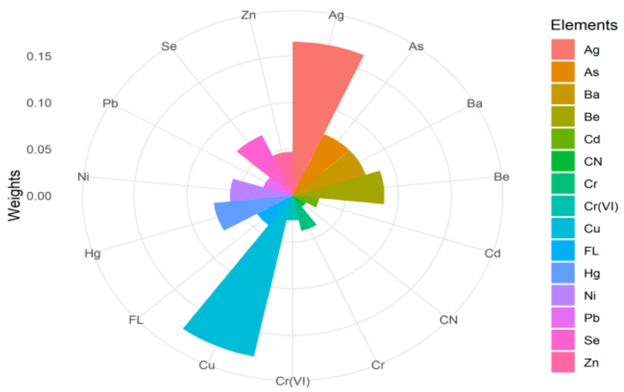


Figure 3. Weighting of toxicity indicators in the calculation process

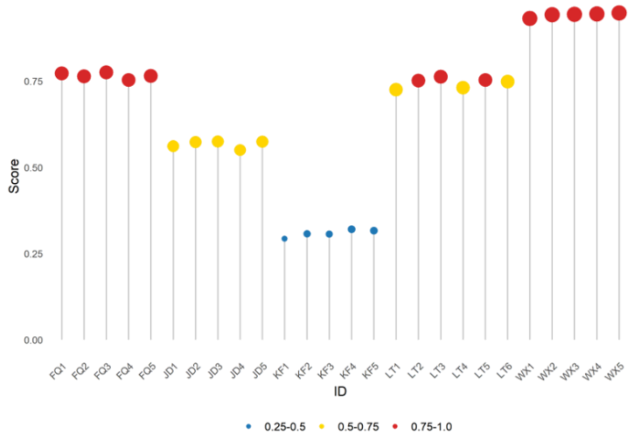


Figure 4. Scores of various mining areas

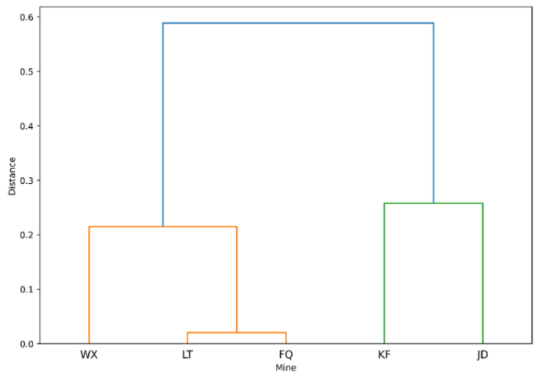


Figure 5. Dendrogram based on hierarchical clustering

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